LIQUEFACTION OF NOVA SCOTIA COALS
PHASE V

STUDIES ON SINGLE-STAGE CATALYTIC HYDROGENATION AND TWO-STAGE THERMAL/CATALYTIC LIQUEFACTION

FINAL REPORT

by

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SUMMARY

The fifth phase of our experimental program on the direct hydroliquefaction of Nova Scotia coal has been completed. This phase of work was concentrated on single-stage catalytic hydrogenation and two-stage thermal/catalytic liquefaction of Lingan Harbour seam coal. Two other coals were tested, Prince Hub and Illinois No. 6. The latter is a highly reactive American coal. During Phase V a total of thirty-eight experiments were conducted in our bench-scale continuous-flow unit. Both thermal and catalytic experiments were carried out in a once-through mode using hydrogenated anthracene oil as vehicle.

Continuous single-stage bench operations have confirmed that catalytic hydrogenation produces higher oil yields than donor solvent extraction. Furthermore, it has been shown that considerable control can be exercised over product distribution by varying process severity. Experiments conducted with Lingan coal and a commercial Ni/Mo hydrotreating catalyst over medium to high severity operating conditions produced high coal conversions, 90-92%, while oil yields ranged from 30-47%. In catalytic hydrogenation experiments with Lingan, Prince and Illinois No. 6 coals at medium severity, 451°C and 17.24 MPa, oil yields were 36, 42 and 57% respectively. A significant difference in reactivity between Illinois No. 6 and Lingan Harbour was also observed at thermal liquefaction conditions. The high reactivity of Illinois No. 6 coal is primarily attributed to its low rank.

During Phase V work began on two-stage thermal/catalytic liquefaction. The process is comprised of a thermal first step to solubilize the coal followed by a catalytic second step to upgrade the first stage product while producing a recycle oil with high donor capacity. Testwork was carried out on both stages separately. First stage dissolution studies at low severity operating conditions have shown that reaction temperature and coal space velocity affect product composition. Non-integrated two-stage trials suggest that high conversions of asphaltenes to oil can be achieved with minimum gas production by reducing slurry space velocity in the catalytic upgrading step. Test results also suggest that a recycle oil of high donor capacity can be produced from Lingan coal.

The next phase of work will concentrate on integrated two-stage liquefaction. A second reactor will be added to the existing unit. The two reactors will be closely coupled to permit integrated two-stage operations. It is expected that extensive scoping of first and second stage operating conditions will be required to optimize selectivity towards distillate products. On completion of the scoping studies we plan to conduct continuous experiments with solvent recycle.

INTRODUCTION

During the past decade several industrialized countries around the world, including Canada, have focused considerable attention on the technical and commercial feasibility of producing liquid fuels from coal. The search for alternate sources of liquid fuels was stimulated by the upward trend in crude oil prices and a delicate supply/demand balance for petroleum products around 1973. In Canada, our way of life is highly dependent on petroleum liquids and should it be economically attractive or essential, abundant coal reserves are available for conversion to synthetic liquid fuels.

Currently the EDS, H-Coal and Ruhrkohle/Veba direct liquefaction processes are at advanced stages of development while synthetic liquid fuels are being produced commercially, under special circumstances in South Africa, by Fisher-Tropsch synthesis. Because long lead times are involved in process evaluation, detailed design and plant construction, it is essential to understand the basic concepts of the promising liquefaction processes and verify their projected yield structures as they apply to Canadian Coals.

Process studies relating to the direct liquefaction of Nova Scotia coals were initiated at the Nova Scotia Research Foundation Corporation in the latter part of 1977. Since that time

five phases of our research program have been completed. Yield data have been developed over a wide range of operating conditions. The product slate of interest in Nova Scotia is liquid transportation fuels. Thus research efforts are directed towards identifying or developing a process configuration to maximize the yield of coal-derived liquids.

The first and second phases of our program were concentrated on producing liquids via low severity extraction/coking (1,2,3). In this approach, hydrogen donor solvent is used under mild conditions(2100 kPa, 400°C) to extract the coal followed by delayed coking as a product separation step. Although low pressure extraction has advantages related to plant hardware there is one major disadvantage, distillate yield is low. Experimental work showed that for one-hour extractions conversions of Lingan coal to benzene solubles plus gases were typically 60%. However, following the coking step distillate yield was ca.30 Wt. %. A variety of methods were investigated to enhance distillate yield from the process but results obtained were not encouraging. Hence it was decided to abandon the low severity approach in pursuit of higher yields of distillate products.

Phases III and IV of the project were concentrated on medium to high pressure donor solvent liquefaction and catalytic hydrogenation. During Phase III, donor solvent extraction experiments were conducted in a high pressure batch system (4). The effects of

temperature, pressure, time and solvent/coal ratio were investigated. Yields of oil, asphaltenes, preasphaltenes and gases were determined. Conversion of Lingan coal increased from 60 to 80% at higher severity operating conditions; 20.68 MPa, 450°C.

During Phase IV a total of ten coals from three operating mines in Cape Breton were tested (5). Continuous donor solvent liquefaction runs were carried out with Lingan, Prince and No. 26 Colliery coals in a once-through mode of operation. The experiments showed that Hub seam coal from the Prince mine is the most reactive feedstock at thermal liquefaction conditions. Within the Harbour seam reactivity was found to decrease in going from the Lingan mine to No. 26. At high severity operating conditions, 460°C and 20.68 MPa, conversions of these coals ranged from 85 - 89% while oil yield ranged from 20 - 34%. Catalytic hydrogenation experiments conducted in our batch autoclave system produced higher oil yields than donor solvent extraction. In tests with Lingan coal the addition of commercial Co/Mo and Ni/Mo hydrotreating catalysts increased oil yield from 26 to 37%. The highest oil yield, 49%, was achieved using a catalyst mixture containing Ni, Mo and Sn.

Phase V of the program began in September of 1983. Actual testwork included an extensive study on single-stage catalytic hydrogenation of Lingan Harbour seam coal. A total of sixteen experiments were conducted in our bench-scale continuous-flow unit. The effects of major process variables on coal conversion and product

distribution were investigated. Details pertaining to operating conditions and product yields are presented herein.

Continuous experiments were also carried out to compare the reactivity of Nova Scotia coals with that of Illinois No. 6, a highly reactive American coal. Direct comparisons were made in both thermal and catalytic modes of operation. The results of these tests are given in Tasks 5 and 6.2.

During Phase V experimental studies were initiated on two-stage thermal/catalytic liquefaction. The two-stage process is comprised of a thermal first step to solubilize the coal followed by a catalytic second step to upgrade the first stage effluent while producing a recycle oil with high donor capacity. Preliminary work was conducted on both stages separately using our existing coal liquefaction unit. Results of first stage dissolution studies and second stage upgrading operations are presented in this report.

TASK 1 - EQUIPMENT MAINTENANCE

During the ten-month period from June 1, 1982 to March 31, 1983 bench unit operations were concentrated on continuous donor solvent liquefaction of Sydney area coals. Prior to initiating the Phase V experimental program one month was taken for inspection and maintenance of the hydrogen compression facility, the fixed bed catalytic hydrotreater and the continuous coal liquefaction unit.

In the hydrogen compression and storage area attention was centered on the Kemlon two-stage compressor. This unit is 3 horsepower with a design capacity of 28 L/min. from 1.5 MPa to 28 MPa. Routine maintenance includes changing the gear oil periodically and adding actuating fluid as required. During this turnaround period process valves were replaced in the second stage and a new stainless steel diaphragm was installed. Minor adjustments were made to the injector and relief valves on the actuating fluid compensator system.

The fixed bed catalytic hydrotreater has been described in an earlier report (3). On turnaround, the reactor was opened and partially spent catalyst was replaced with fresh material. New packing rings were installed in the liquid metering pump and heating tapes were replaced on feed and product transfer lines. Additionally, the heated feed vessel was examined and deteriorating insulation was replaced.

A complete description of the continuous-flow coal liquefaction unit is presented elsewhere (5). This system was used

extensively during the first quarter of 1983 without encountering any significant mechanical problems. Most of the actual work associated with Task 1 was devoted to the inspection and maintenance of this unit.

The first step was a general overhaul of the slurry circulation and high pressure metering pumps. Then an orderly disassembly of the rig began. The slurry circulation loop was taken apart, cleaned and new ball valves were installed below the feed tank. Next, slurry feed and product transfer lines were disassembled. The transfer lines and associated fittings were found to be essentially free of solids. However, on removing the bottom cover of the stirred tank reactor it was found that solids, saturated with oil, had deposited on and near baffles contacting the reactor wall and at the bottom of the vessel.

The gummy deposits were collected as two separate samples. In both cases the material was easily removed. The individual samples were washed with an excess of pentane to remove the oil and then dried. Approximately 31g of dry solids were recovered, 25g from the reactor wall and baffles and 6g from the bottom. On further examination, solids from the wall/baffle deposit were found to contain 12.1% volatile matter, 20.7% fixed carbon and 67.2% ash. Solids from the bottom deposit contained 12.4% volatile matter, 7.2% fixed carbon and 80.4% ash. It is suggested that the deposition of solids is specifically related to the internal geometry of the reactor and does not significantly affect test results.

A thorough inspection was carried out on all high pressure valves. In order to reduce maintenance requirements for valves in hot slurry service graphite-based packing was substituted for teflon. Finally, deteriorated stellite stems and seats in the manual product let-down valves were replaced.

As mentioned elsewhere (5) gamma ray equipment designed to measure and control liquid level in the primary high pressure separator was installed during the preceding phase of work. Unfortunately, the system did not function properly due to a defective amplifier supplied with the Kay-Ray ion chamber detector. Lengthy delays were encountered in testing the electronics and obtaining the required new components. However, repairs were completed on the amplifier by September of 1983 and the automated high pressure let-down system was used successfully throughout Phase V.

TASK 2 - PREPARATION AND ANALYSIS OF MATERIALS

2.1 Coal Feedstocks

Three coal feedstocks were tested during this phase of the project. The experimental program was principally focused on Lingan Harbour seam coal with less emphasis on Prince Hub seam and Illinois No. 6 from the Burning Star Mine. Lingan and Prince mine coals were provided courtesy of the Cape Breton Development Corporation. Illinois No. 6 was procured through Cepheus Industries Incorporated, Marion, Illinois. Preparation involved riffling, grinding and screening. After screening the individual coals were thoroughly mixed and stored in plastic-lined drums with a nitrogen blanket.

Proximate and ultimate analysis of the coals are given in Table 1. Proximate analysis, free swelling index and forms of sulphur were determined according to standard methods. Total sulphur was measured using Eschka's method. Carbon, hydrogen and nitrogen were determined using a Perkin-Elmer elemental analyzer.

Petrographic analyses of the three coals are given in Table 2. These analyses were carried out by coal petrology experts at the Bedford Institute of Oceanography.

2.2 Anthracene Oil

This material is a coal-tar distillate which contains ca. 5.6 Wt. % hydrogen. It was supplied by Domtar Chemicals Limited (Hamilton, Ontario). Hydrogenated solvents used in the continuous coal liquefaction experiments were made from two batches of this highly aromatic oil.

TABLE 1

ANALYSIS OF COALS TESTED

COAL DESCRIPTION:	LINGAN BATCH L	PRINCE BATCH D	ILLINOIS NO. 6
Moisture, Wt. %	1.28	2.45	3.17
PROXIMATE (Dry Basis) Wt. %			
Ash	4.10	9.23	11.13
Volatile Matter	34.83	36.22	38.05
ULTIMATE (Dry Basis) Wt. %			
Carbon	81.78	73.48	68.73
Hydrogen	5.37	4.80	4.73
Nitrogen	1.56	1.45	1.39
Sulphur	1.67	4.22	3.46
Ash	4.10	9.23	11.13
Oxygen (by difference)	5.85	6.82	10.56
SULPHUR FORMS (Dry Basis) Wt. %			
Sulphate	0.05	0.18	0.01
Pyritic	1.05	2.25	1.50
Organic (by difference)	0.57	1.79	1.95
Total	1.67	4.22	3.46
FREE SWELLING INDEX	8	2	1½

TABLE 2

PETROGRAPHIC CHARACTERISTICS OF COALS TESTED

COAL DESCRIPTION:	LINGAN BATCH L	PRINCE BATCH D	ILLINOIS NO. 6
MACERAL COMPOSITION (% by Volume)			
Vitrinite	77.0	74.7	79.2
Exinite	4.2	4.1	3.8
Inertinite	16.4	18.0	11.8
Macrinite	(2.6)	(0.8)	(0.6)
Micrinite	(4.4)	(5.4)	(4.6)
Semifusinite	(4.6)	(5.4)	(2.8)
Fusinite	(4.8)	(6.4)	(3.8)
Mineral Matter	2.4	3.2	5.2
Pyrite	(0.6)	(2.6)	(1.4)
Quartz	(0.4)	- -	(1.4)
Shale	(1.4)	(0.4)	(2.4)
Calcite	- -	(0.2)	- -
% R_o Max.	0.96	0.78	0.62

The characteristics of the raw oil are presented in Table 3. Carbon and hydrogen were determined using a Perkin-Elmer elemental analyzer. A standard Kjeldahl method was used for the determination of nitrogen. Total sulphur content was measured by X-ray fluorescence. Oxygen was determined using a Carlo Erba elemental analyzer.

In this study the hydrogen aromatic/aliphatic distribution was established by proton nuclear magnetic resonance (H-NMR). A Varian CFT-20 NMR spectrometer operating in the Fourier transform mode at 80 MHz and a Nicolet 360NB NMR spectrometer operating in the Fourier transform mode at 360 MHz were used to obtain H-NMR spectra. Proton assignments were as follows: aromatic hydrogens (H_{ar}) are found in the region of 9-6ppm. Benzylic hydrogens, including methyls, occur at 3.5 to 2 ppm (H_{α}). Hydrogens, except methyls, on carbons one or more positions removed from aromatic rings, occur at 2-1.1 ppm (H_{β}). Aliphatic methyls are found at 1.1 - 0.4 ppm (H_{γ}). As almost no long aliphatic side chains are believed to be present, the H_{β} protons are predominantly in rings. These assignments are consistent with similar analyses reported in the literature (6,7). Figure 1 is a typical H-NMR spectrum of raw anthracene oil.

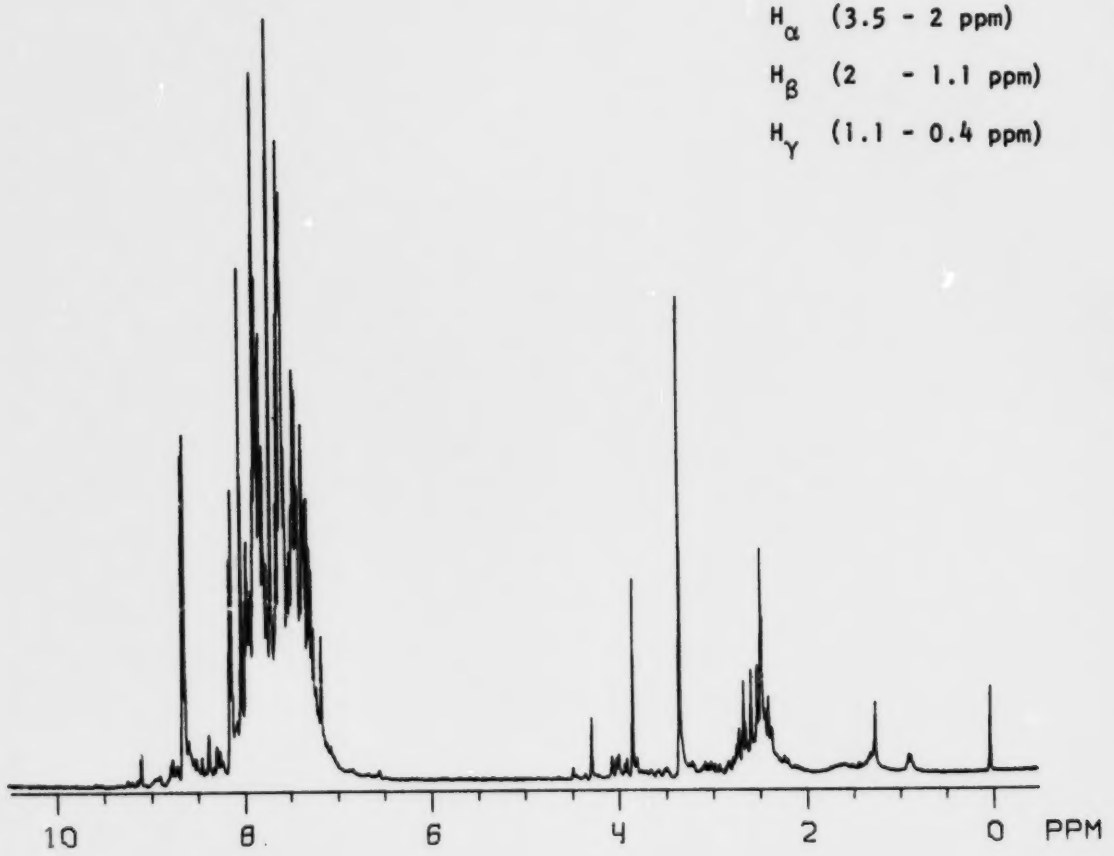
TABLE 3

ANALYSIS OF RAW ANTHRACENE OIL

SAMPLE DESCRIPTION	BATCH NO.4	BATCH NO.5
Specific Gravity, 60/60°F	1.135	1.152
Carbon, Wt. %	91.03	91.67
Hydrogen, Wt. %	5.65	5.62
Nitrogen, Wt. %	0.97	0.88
Sulphur, Wt. %	0.62	0.56
Oxygen, Wt. %	1.29	1.21
Proton Distribution, %		
H_{ar} ((9-6 ppm)	86.21	86.87
H_{α} (3.5 - 2 ppm)	11.30	10.74
H_{β} (2 - 1.1 ppm)	1.48	0.80
H_{γ} (1.1 - 0.4 ppm)	1.01	0.80
Distillation Data, °C (ASTM D-1160)		
IBP	227	223
10%	270	291
20%	301	316
30%	320	331
40%	336	346
50%	348	355
60%	362	367
70%	375	382
80%	390	399
90%	416	434
95%	455	461
EP	470	482

Figure 1 - H-NMR SPECTRA OF RAW ANTHRACENE OIL WITH 5.65 WT. % HYDROGEN

PROTON DISTRIBUTION, %	
H_{ar} (9 - 6 ppm)	86.21
H_{α} (3.5 - 2 ppm)	11.30
H_{β} (2 - 1.1 ppm)	1.48
H_{γ} (1.1 - 0.4 ppm)	1.01



2.3 Catalysts

Two commercially available hydrotreating catalysts, Nalco NM 502 and American Cyanamid HDS-1442 were evaluated in the catalytic coal hydrogenation experimental program. The former catalyst is alumina-supported $NiO-MoO_3$ whereas the latter is alumina-supported $CoO-MoO_3$. Both catalysts are manufactured in extrudate form, 1.6 mm diameter. Because catalyst was used in a once-through mode during this phase of work these materials were ground to -100 mesh prior to slurry preparation. Nalcomo 479 was used as received in the fixed-bed catalytic hydrotreater. The chemical and physical properties of these Ni/Mo and Co/Mo catalysts are given in Table 4. This data was provided by the manufacturers.

TABLE 4

CHEMICAL AND PHYSICAL PROPERTIES OF HYDROTREATING CATALYSTS

	NALCO NM 502	HDS-1442	NALCOMO 479
MoO_3, Wt. %	14.0	13.2	19.0
NiO, Wt. %	4.0		
CoO, Wt. %		3.5	4.4
Size, cm	0.16	0.16	0.16
Surface Area, M^2/g	240	300	180
Pore Volume, cc/g	0.53	0.80	0.47
Density, g/cc	0.73 - 0.74	0.58	0.82 - 0.84
Crush Strength, kg	6.4 - 7.3	6.2	6.4 - 7.3

Nalco NM 502 and Nalcomo 479 are manufactured by the Nalco Chemical Company.

HDS-1442 is manufactured by American Cyanamid.

TASK 3 - HYDROGENATION OF ANTHRACENE OIL

This task was an essential part of Phase V. Hydrogenated solvents were prepared from raw anthracene oil using a fixed-bed catalytic hydrotreater. The bench-scale unit has been described in a previous report (3).

A summary of the operating conditions used to produce solvents containing 7.5 and 9.0% hydrogen is given in Table 5. The characteristics of the two hydrogenated oils are presented in Table 6. A comparison of distillation data (ASTM D-1160) is shown in Figure 2. H-NMR spectra for the 7.5 and 9.0% H solvents are given in Figures 3 and 4.

The continuous coal liquefaction experiments described in Tasks 5, 6 and 7 required in excess of 350 liters of hydrogenated solvent. Fortunately, there was a sufficient quantity of donor solvent, 9% H, on-hand to conduct Task 5. The additional requirement of 9% H oil was prepared at the rate of 1.6 liters per 7-hour man-day whereas 7.5% H oil was produced at twice that rate. In both cases daily production was collected to obtain 20-liter batches. A total of fifteen 20-liter batches were prepared, eight of which were 7.5% H oil. Analysis of selected batches are given in Tables 7 and 8. Prior to initiating Task 6 the individual 20-liter batches of 7.5% H oil were blended. One large batch of 9% H oil was also prepared in a similar fashion before starting Task 7. The analysis presented in Table 6 are representative of the 7.5 and 9% H oil blends.

TABLE 5

REACTION CONDITIONS FOR ANTHRACENE OIL HYDROGENATION

Reactor Temperature, ^{o}C	368	375
Reactor Pressure, MPa	13.79	15.51
LHSV, hr^{-1}	0.64-0.76	0.20
Hydrogen Flow, L/hr	225 - 250	125 - 225
Product Hydrogen Content, Wt. %	7.54	9.01

Catalyst used: Nalcomo 479

TABLE 6

CHARACTERISTICS OF HYDROGENATED ANTHRACENE OIL

SAMPLE DESCRIPTION	HAO 55	HAO 56
Specific Gravity, 60/60°F	1.073	1.033
Carbon, Wt. %	91.58	90.36
Hydrogen, Wt. %	7.54	9.01
Nitrogen, Wt. %	0.41	0.13
Sulphur, Wt. %	0.062	0.006
Oxygen, Wt. %	0.96	1.06
Proton Distribution, %		
H_{ar} (9-6 ppm)	36.49	16.34
H_{α} (3.5 - 2 ppm)	31.75	33.26
H_{β} (2 - 1.1 ppm)	24.57	37.77
H_{γ} (1.1 - 0.4 ppm)	7.18	12.63
Distillation Data, °C (ASTM D-1160)		
IBP	188	177
10%	251	251
20%	282	277
30%	300	294
40%	312	305
50%	323	314
60%	334	323
70%	347	336
80%	366	356
90%	387	384
95%	404	405
EP	438	437

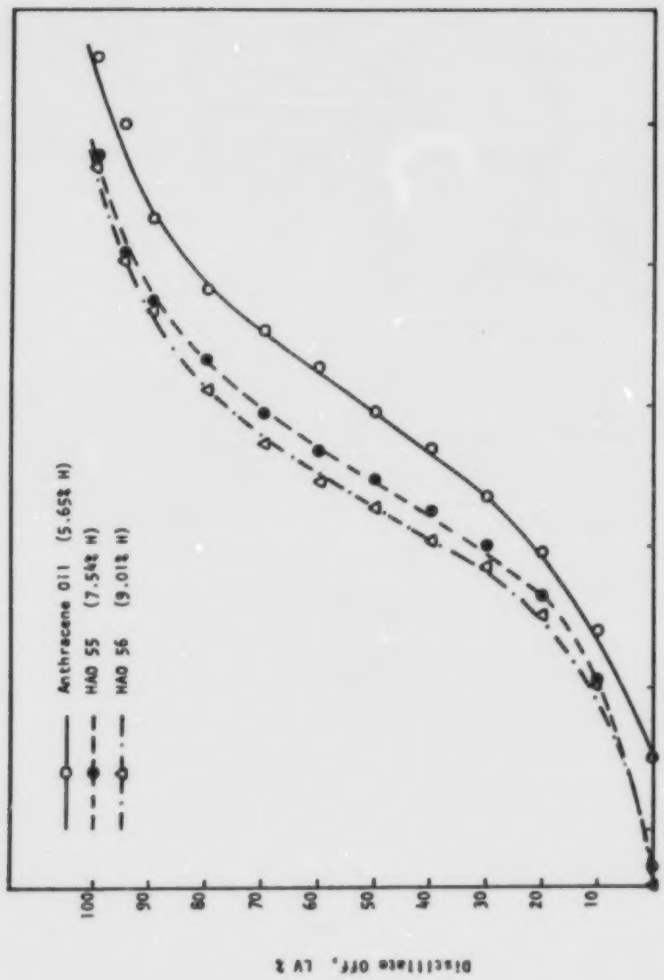


Figure 2 - COMPARISON OF ASTM DISTILLATIONS (D-1160) FOR RAW AND HYDROGENATED ANTHRACENE OIL

Figure 3 - H-NMR SPECTRA OF HYDROGENATED ANTHRACENE OIL WITH 7.54 WT. % HYDROGEN

PROTON DISTRIBUTION, %		
H_{ar}	(9 - 6 ppm)	36.49
H_{α}	(3.5 - 2 ppm)	31.75
H_{β}	(2 - 1.1 ppm)	24.57
H_{γ}	(1.1 - 0.4 ppm)	7.18

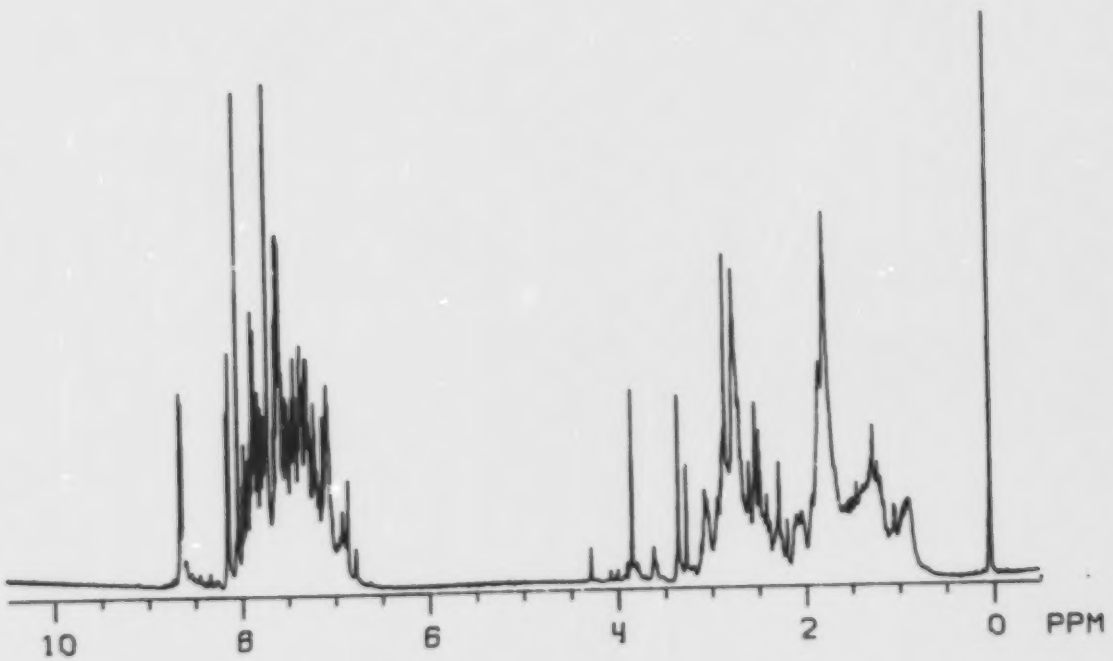


Figure 4 - H-NMR SPECTRA OF HYDROGENATED ANTHRACENE OIL WITH 9.01 WT. % HYDROGEN

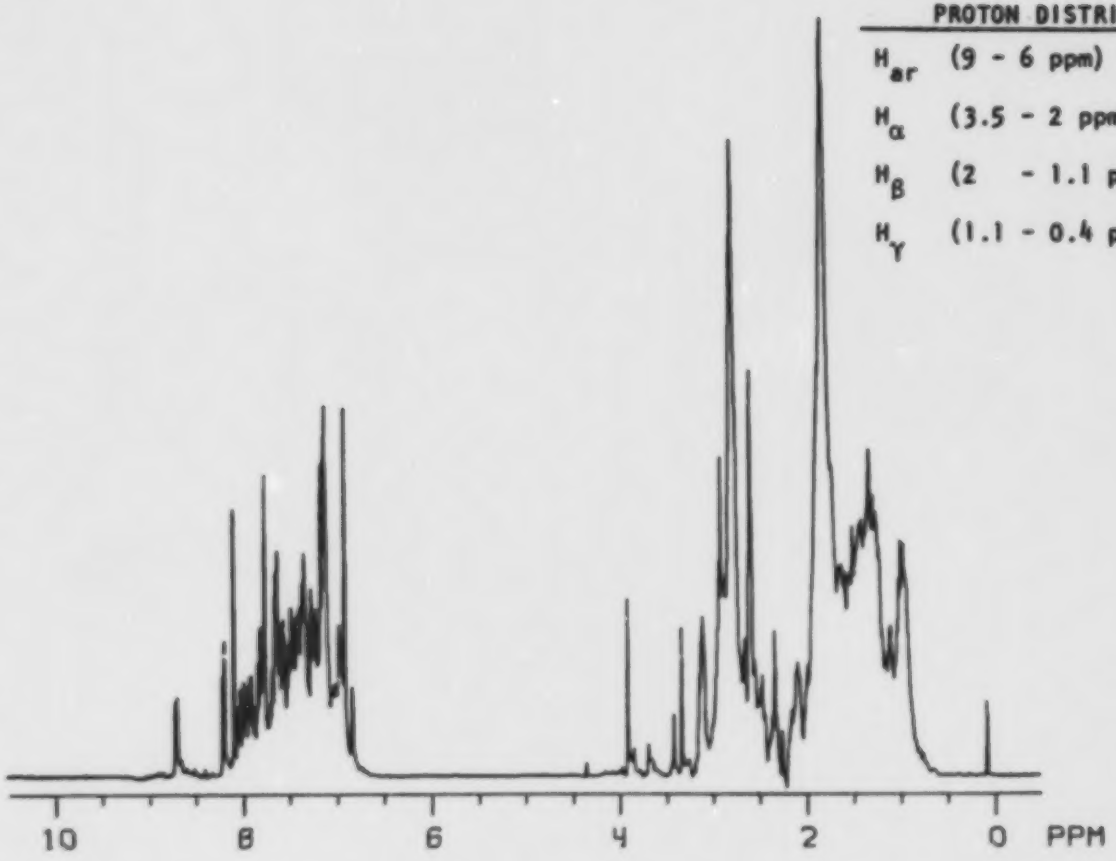


TABLE 7

ANALYSIS OF 20-LITER BATCHES OF 7.5 % H SOLVENT

BATCH NO.	1	2	3	4	5	6	7	8
Specific Gravity, 60/60°F	1.076	1.076	1.073	1.073	1.073	1.071	1.071	1.071
Carbon, Wt. %		90.79			90.81			92.92
Hydrogen, Wt. %		7.55			7.45			7.55
Nitrogen, Wt. %		0.41			0.40			0.37
Sulphur, Wt. %		0.063			0.051			0.049
Proton Distribution, %								
H_{ar} (9-6 ppm)		37.15			36.16			33.63
H_{α} (3.5 - 2 ppm)		31.95			35.41			33.23
H_{β} (2 - 1.1 ppm)		24.17			23.75			26.52
H_{γ} (1.1 - 0.4 ppm)		6.73			4.68			6.63
Distillation Data, °C (ASTM D-86)								
IBP	188	195	192	186	160	196	190	182
5%	255	250	252	255	248	248	248	243
10%	273	268	270	264	267	271	270	262
20%	290	286	288	287	284	287	291	287
30%	303	300	304	302	301	302	306	302
40%	316	316	315	314	316	315	317	315
50%	331	328	328	327	328	328	330	328
60%	342	340	340	337	340	338	342	340
70%	357	353	352	350	352	352	355	352
80%	375	370	368	367	370	368	370	371
90%	393	393	390	390	391	391	390	390
95%	410	410	406	406	398	409	400	408
EP	493	>500	498	482	481	438	>500	500

TABLE 8

ANALYSIS OF 20-LITER BATCHES OF 9% H SOLVENT

BATCH NO.	9	10	11	12	13	14	15
Specific Gravity, 60/60°F	1.025	1.028	1.030	1.031	1.031	1.036	1.038
Carbon, Wt. %	89.41	89.66	90.60	90.06	90.05	89.58	89.51
Hydrogen, Wt. %	9.09	9.19	9.00	8.96	9.06	8.80	8.78
Nitrogen, Wt. %	0.09		0.13		0.10		0.13
Sulphur, Wt. %	0.005		0.007		0.006		0.005
Proton Distribution, %							
H_{ar} (9-6 ppm)	16.37		17.08			17.61	
H_{α} (3.5 - 2 ppm)	32.22		33.78			35.08	
H_{β} (2 - 1.1 ppm)	38.69		38.07			37.21	
H_{γ} (1.1 - 0.4 ppm)	12.72		11.07			10.10	
Distillation Data, °C (ASTM D-86)							
IBP	185	182	185	175	176	177	172
5%	241	242	240	242	258	256	253
10%	263	257	260	265	273	275	270
20%	278	278	280	282	288	293	288
30%	293	291	293	295	300	306	299
40%	305	301	303	307	311	316	310
50%	316	313	315	320	320	327	321
60%	328	325	326	330	330	336	331
70%	339	337	338	341	342	348	344
80%	354	353	356	356	357	363	362
90%	378	376	380	381	382	389	389
95%	403	395	401	399	401	409	414
EP	>500	>500	490	484	496	>500	>500

TASK 4 - PROCUREMENT AND INSTALLATION OF NEW EQUIPMENT

The purpose of Task 4 was to upgrade our coal liquefaction facility. Activities under this task included procurement of equipment and assembly of a batch vacuum distillation system for product separation.

Major pieces of equipment acquired to improve the operability and expand the flexibility of the coal liquefaction unit were as follows:

(1) Bran and Lubbe slurry metering pump with two pumpheads.

(2) Air-operated valves and associated high pressure hardware required for a second automatic product let-down circuit.

(3) Stirred tank reactor, 3.8-liter capacity, complete with furnace to be used as the catalytic reaction zone in our proposed integrated two-stage testwork. Although this reactor was not required for Phase V it was purchased because of the long lead time associated with its delivery.

The original plan was to install the Bran and Lubbe pump and the backup automatic product let-down circuit during Phase V. However, there were delays in receiving equipment and the coal liquefaction unit was operating very well. Therefore, installation of this equipment was postponed until Phase VI. At the start of Phase VI the

coal liquefaction unit will be modified for integrated two-stage operations.

Vacuum distillation is commonly used for product separation in coal liquefaction. The procurement of a laboratory scale vacuum distillation system to be used primarily for solvent recycle liquefaction experiments was part of Task 4. Commercial suppliers of batch and continuous systems were contacted. It was found that the systems offered were either too fragile or there was a high level of risk associated with mechanical operability. In consideration of these findings a batch system was designed and built at NSRFC.

A schematic of the distillation apparatus is shown in Figure 5. The vessels, transfer lines and associated fittings are made of type 316 stainless steel. The system is comprised of a nominal 20-liter distillation flask, an electric heating mantle, two product receivers, two knock-out vessels, a condenser, a demister, temperature measuring devices, a mercury manometer and vacuum control equipment. The system is assembled on an open steel framework 152 cm L x 91 cm W x 214 cm H.

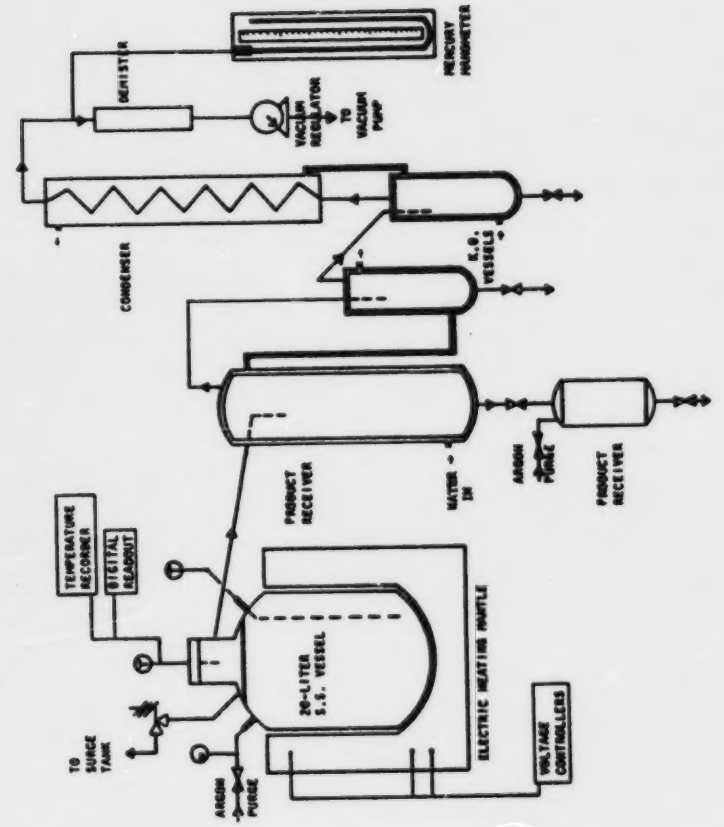


Figure 5 - SIMPLIFIED FLOW DIAGRAM OF 20-LITER BATCH VACUUM DISTILLATION UNIT

TASK 5 - DONOR SOLVENT LIQUEFACTION

Coal liquefaction process developers in countries around the world have used Illinois No. 6 coal as a benchmark feedstock. The objective of Task 5 was to make a direct comparison of the thermal reactivity of Nova Scotia coal with that of Illinois No. 6. This was accomplished by running Lingan Harbour seam and Illinois No. 6 coals in our bench unit under donor solvent liquefaction conditions.

The continuous-flow bench unit has been described elsewhere (5). Duplicate runs were carried out on each coal using hydrogenated anthracene oil containing 9% H as the donor vehicle. The concentration of coal in the feed slurry was fixed at 30 Wt. %. Hydrogen treatrate was ca. 7.5 Wt. % based on dry coal. Other operating conditions were as follows:

Reactor Temperature, ^{o}C	460
Back Pressure, MPa	20.68
Slurry Feedrate, kg/hr	2

At a slurry feedrate of 2 kg/hr the average residence time of coal particles in the reaction zone is estimated to be 21 minutes. This estimate is based on the physical properties of the vehicle oil at operating conditions. The physical data required was developed experimentally.

Each donor solvent liquefaction run was of six hours duration. However, an additional 5-6 hours was required for preliminary

and post run operations. In this series of runs the controlled variables lined-out about 4 hours after start-up. During the steady state operating period vent gas streams were sampled and other product streams were collected for analytical and material balance purposes. A detailed operating procedure is given in Appendix I.

In all Phase V experiments the following definitions for the product fractions isolated by solvent extraction were used:

Oil - pentane solubles

Asphaltenes - toluene solubles, pentane insolubles.

Preasphaltenes - THF solubles, toluene insolubles.

A direct extraction technique was used to determine these fractions. Individual samples of product slurry were separately extracted with pentane, toluene and tetrahydrofuran (THF). Conversions were calculated on the basis of maf coal fed to the reactor during the steady state sampling period and unconverted coal in the dry insoluble residues. Conversion calculations are described in Appendix A.

Vent gas streams were analyzed using a Perkin-Elmer 3B gas chromatograph interfaced with a Perkin-Elmer Sigma 10 Chromatography data station. Simulated distillations were also done with this instrumentation using the D-2887 Sigma Basic program. Distillation data for pentane and toluene solubles are given in Appendix D.

Results of the continuous donor solvent liquefaction experiments are presented in Table 9. Coal conversion averaged 92.9% in Run Nos. 5001 and 5002 with Illinois No. 6. Product distribution

TABLE 9

DONOR SOLVENT LIQUEFACTION OF LINGAN AND ILLINOIS NO. 6 COALS

RUN NO. COAL TESTED	5001 ILLINOIS NO. 6	5002 ILLINOIS NO. 6	5003 LINGAN BATCH L	5004 LINGAN BATCH L
Temperature, °C	460	460	460	460
Back Pressure, MPa	20.68	20.68	20.68	20.68
Slurry Feedrate, kg/hr	2.048	2.015	2.059	2.003
H_2 Consumption, Wt. % maf Coal	1.0	0.7	0.9	1.3
CONVERSIONS, WT. % OF maf COAL				
Pentane Solubles plus Gases	47.6	47.3	33.5	35.0
Toluene Solubles plus Gases	75.0	77.3	62.7	64.1
THF Solubles plus Gases	92.6	93.1	86.0	86.3
PRODUCT DISTRIBUTION, WT. % OF maf COAL				
Gases	17.8	18.6	12.0	12.9
Oil	29.8	28.7	21.5	22.1
Asphaltenes	27.4	30.0	29.2	29.1
Preasphaltenes	17.6	15.8	23.3	22.2

Coal concentration in feed slurry; 30 Wt. %.
Detailed product distribution in Appendix B.

averaged 18.2% gases, 29.3% oil, 28.7% asphaltenes and 16.7% preasphaltenes. In Run Nos. 5003 and 5004 with Lingan Harbour coal, conversions were 86.0 and 86.3% respectively. Product yields averaged 12.5% gases, 21.8% oil, 29.2% asphaltenes and 22.8% preasphaltenes. In summary, Run Nos. 5001 and 5002 produced higher coal conversions, higher yields of light products and less preasphaltenes than Run Nos. 5003 and 5004. This data illustrates that Illinois No. 6 coal is more reactive than Lingan Harbour at thermal liquefaction conditions.

The coals tested differ in both their chemical and petrographic characteristics as shown in Tables 1 and 2. Illinois No. 6 is a lower rank coal and has higher levels of ash and total sulphur than Lingan Harbour. These specific properties are known to influence the liquefaction behavior of coal. However, based on our earlier experimental work and knowledge from published literature, the difference in reactivity found here is primarily attributable to coal rank. Furthermore, the lower level of inertinite components in Illinois No. 6 favours higher coal conversion.

TASK 6 - CATALYTIC HYDROGENATION

The purpose of this task was to conduct catalytic hydroliquefaction studies on Sydney area coals in a continuous-flow bench unit. The test program was primarily focused on Lingan Harbour seam coal with less emphasis on Hub seam coal from the Prince mine. Product yield structures were established for Lingan Harbour, Prince Hub and Illinois No. 6 at similar operating conditions to extend the comparison of coal reactivity initiated in Task 5.

In this study pulverized catalyst was added to the feed slurry to simulate single-stage catalytic coal liquefaction. The selected coals were processed in a once-through mode using hydrogenated anthracene oil as vehicle.

6.1 Process Variable Study on Lingan Harbour Seam Coal

A total of eleven experiments were carried out on Lingan Harbour coal at medium severity operating conditions to investigate conversion and product yields. The experiments were conducted varying reaction temperature, coal space velocity, hydrogen treatrate, vehicle oil composition, catalyst type and catalyst concentration. Runs were of 6 - 7 hours duration. A summary of operating conditions and results are shown in Table 10.

The first three experiments, Run Nos. 5005 - 5007, were carried out at 460°C and 17.24 MPa. The concentration of coal in the feed slurry was fixed at 29.5 Wt. %. A commercially available Ni/Mo hydrotreating catalyst, Nalco NM 502, was used. In Run No. 5005

TABLE 10

CATALYTIC HYDROGENATION OF LINGAN COAL AT MEDIUM SEVERITY OPERATING CONDITIONS

RUN NO.	5005	5006	5007	5009	5010	5011	5012	5014
Temperature, °C	460	460	460	451	451	451	467	455
Back Pressure, MPa	17.24	17.24	17.24	17.24	17.24	17.24	17.24	17.24
Slurry Feedrate, kg/hr	1.054	1.009	0.793	0.925	0.925	0.942	0.927	1.319
H_2 Treatrate, Wt. % Dry Coal	7.5	8.0	15.1	11.0	14.9	12.8	13.0	13.0
Catalyst Level, Wt. % maf Coal	4	8	8	7	7	10	8	8
H_2 Consumption, Wt. % maf Coal	4.0	4.3	5.6	4.1	4.8	4.0	4.8	4.3
CONVERSIONS, WT. % OF maf COAL								
Pentane Solubles plus Gases	45.0	50.2	60.1	50.7	51.2	52.5	59.7	49.0
Toluene Solubles plus Gases	76.8	79.8	84.6	79.7	79.5	80.6	84.9	77.0
THF Solubles plus Gases	91.0	90.9	91.6	91.4	91.6	91.2	92.1	90.5
PRODUCT DISTRIBUTION, WT. % OF maf COAL								
Gases	15.1	17.1	21.9	14.2	16.5	16.4	25.1	15.9
Oil	29.9	33.1	38.2	36.5	34.7	36.1	34.6	33.1
Asphaltenes	31.8	29.6	24.5	29.0	28.3	28.1	25.2	28.0
Preasphaltenes	14.2	11.1	7.0	11.7	12.1	10.6	7.2	13.5

TABLE 10 (Continued)

CATALYTIC HYDROGENATION OF LINGAN COAL AT MEDIUM SEVERITY OPERATING CONDITIONS

RUN NO.	5015	5016	5017
Temperature, ^{o}C	455	460	460
Back Pressure, MPa	17.24	17.24	17.24
Slurry Feedrate, kg/hr	0.926	0.858	0.862
H_2 Treatrate, Wt. % Dry Coal	12.0	14.0	12.2
Catalyst Level, Wt. % maf Coal	8	8	8
H_2 Consumption, Wt. % maf Coal	4.1	4.4	3.7
CONVERSIONS, WT. % OF maf COAL			
Pentane Solubles plus Gases	52.6	55.9	60.9
Toluene Solubles plus Gases	80.7	81.6	83.8
THF Solubles plus Gases	90.7	90.9	91.9
PRODUCT DISTRIBUTION, WT. % OF maf COAL			
Gases	18.3	22.0	22.7
Oil	34.3	33.9	38.2
Asphaltenes	28.1	25.7	22.9
Preasphaltenes	10.0	9.3	8.1

Coal concentration in feed slurry; 29.5 Wt. %.
Nalco NM 502 catalyst used in Run Nos. 5005-5015 and 5017.
HDS-1442 catalyst used in Run No. 5016.
7.5% H vehicle oil used in Run Nos. 5005-5016.
9.0% H vehicle oil used in Run No. 5017.
Detailed product distribution given in Appendix B.

the slurry feedrate was 1 kg/hr. Catalyst concentration was fixed at 4 Wt. % based on maf coal and hydrogen treatrate was 7.5 Wt. % based on dry coal. The coal conversion and oil yield produced were 91.0 and 29.9% respectively. In Run No. 5006 catalyst concentration was doubled to 8 Wt. %. Coal conversion to THF solubles plus gases was similar to that of Run No. 5005, however, oil yield increased to 33.1%. In Run No. 5007 catalyst was held at the 8 Wt.% level, hydrogen treatrate was doubled to 15 Wt. % and slurry feedrate was reduced to 0.8 kg/hr. The changes in hydrogen treatrate and slurry feedrate affected a significant shift in product distribution. Oil yield increased further to 38.2% while preasphaltenes decreased to 7.0%. The gas make in Run No. 5007 was 21.9% compared with 17.1% in Run No. 5006.

On completion of these baseline runs an optimization study was undertaken. Five experiments were carried out according to a Simplex Self-Directing Evolutionary Operation (SSEVOP). The primary goal of SSEVOP is to locate the optimum operating conditions as quickly as possible when several variables are being considered. In this case temperature, catalyst level, hydrogen treatrate and slurry feedrate were the process variables of interest. The principal response was oil yield.

An analysis of operating data was made to determine the average level and range for each factor. It was judged that a shift of one standard deviation from the mean level in each direction would be

acceptable for each factor. The original Simplex design was prepared based on the results of this preliminary analysis. Details are shown in Appendix G. The run conditions established were as follows:

RUN NO.	HYDROGEN TREATRATE (WT. % DRY COAL)	CATALYST LEVEL (WT. % maf COAL)	TEMPERATURE °C	SLURRY FEEDRATE (kg/hr)
5009	11	7	451	0.920
5010	15	7	451	0.920
5011	13	10	451	0.920
5012	13	8	467	0.920
5014	13	8	455	1.320

Test results are presented in Table 10. Oil yield ranged from 33.1 to 36.5%. The results of these five runs were used to calculate a new design point (see Appendix G).

The operating conditions at the new design point were 455°C, 8% catalyst, 13% hydrogen treatrate and 0.5 kg/hr slurry feedrate. These conditions were similar to those used in Run No. 5014 with the exception of slurry feedrate. The new design point could not be run however, because previous attempts to operate the metering pump at this level were unsuccessful. In consideration of these facts Run No. 5015 was conducted at a slurry feedrate of 0.9 kg/hr, the mid-point between the new design point and the feedrate used in Run No. 5014. A comparison of results from Run Nos. 5014 and 5015 shows that the decrease in slurry feedrate from 1.3 to 0.9 kg/hr did not affect oil yield substantially.

At this point two runs were carried out to investigate catalyst type and vehicle oil composition. American Cyanamid HDS-1442, a Co/Mo hydrotreating catalyst was used in Run No. 5016. A comparison of Run Nos. 5007 and 5016 shows a higher oil yield with the Ni/Mo catalyst. These results are in agreement with our earlier findings using a batch autoclave system for catalyst screening tests.

Hydrogenated anthracene oil containing 7.5 Wt. % hydrogen was used as vehicle oil in Run Nos. 5005 - 5016 inclusive. In Run No. 5017 hydrogenated anthracene oil with a hydrogen content of 9% was used. A comparison of product yields from Run Nos. 5015 and 5017 shows that a small improvement in oil yield was achieved using 9% H vehicle oil. The highest coal conversion and oil yield achieved at medium severity operating conditions were 92 and 38% respectively.

6.2 Comparison of Lingan Harbour, Prince Hub and Illinois No. 6 Coal Reactivity

A total of five runs are discussed in this section. Duplicate runs conducted with Prince Hub and Illinois No. 6 are compared with a single run on Lingan Harbour coal. The operating conditions were as follows: 451°C, 17.24 MPa, 13% hydrogen treatrate and 0.92 kg/hr slurry feedrate. Nalco NM 502 catalyst was used at a 10 Wt. % level. A summary of test results are presented in Table 11.

An examination of the data shows that high levels of conversion to THF solubles plus gases were achieved with all three coal feedstocks. In fact, total coal conversion ranged from 90.5 to 93.6%. However, product distributions were significantly different.

TABLE II

CONVERSION AND PRODUCT DISTRIBUTION DATA FOR LINGAN, PRINCE AND ILLINOIS NO. 6 COALS

RUN NO. COAL USED	5011 LINGAN BATCH L	5018 ILLINOIS NO. 6	5019 ILLINOIS NO. 6	5020 PRINCE BATCH D	5020A PRINCE BATCH D
Temperature, °C	451	451	451	451	451
Back Pressure, MPa	17.24	17.24	17.24	17.24	17.24
Slurry Feedrate, kg/hr	0.942	0.936	0.953	0.922	0.915
H_2 Treatrate, Wt. % Dry Coal	12.8	13.1	13.0	13.2	13.0
Catalyst Level, Wt. % maf Coal	10	10	10	10	10
H_2 Consumption, Wt. % maf Coal	4.0	5.9	5.7	4.6	5.1
CONVERSIONS, WT. % OF maf COAL					
Pentane Solubles plus Gases	52.5	78.0	76.9	63.4	61.8
Toluene Solubles plus Gases	80.6	91.9	91.7	85.6	85.9
THF Solubles plus Gases	91.2	93.6	93.3	90.6	90.5
PRODUCT DISTRIBUTION, WT. % OF maf COAL					
Gases	16.4	20.3	20.1	20.9	20.8
Oil	36.1	57.7	56.8	42.5	41.0
Asphaltenes	28.1	13.9	14.8	22.2	24.1
Preasphaltenes	10.6	1.7	1.6	5.0	4.6

Vehicle oil used: hydrogenated anthracene oil, 7.5% H.
Coal concentration in feed slurry; 29.5 Wt. %.
Detailed product distribution given in Appendix B.

For example, Run No. 5011 with Lingan coal produced 36.1% oil whereas the oil yields for Prince and Illinois No. 6 averaged 41.8 and 57.3% respectively. Furthermore, the results show that in the case of the Nova Scotia coals the conversion products contain high levels of asphaltenes and preasphaltenes. The asphaltene/preasphaltene components in Lingan raw product totalled 38.7%. This is more than double the 16% yield of these components measured in Illinois No. 6 raw product.

These experiments accentuate the difference in reactivity observed between Lingan Harbour and Illinois No. 6 at thermal liquefaction conditions. As stated in Task 5 these coals differ chemically and petrographically. With regard to chemical characteristics Lingan has lower ash and sulphur levels than Illinois No. 6. Prince Hub, on the other hand, has high ash and sulphur levels similar to those of Illinois No. 6. Figures 6 - 10 are presented to illustrate the variation of conversion with individual coal properties.

Figure 6 suggests that ash content has a positive influence on product distribution. However, since there are only three data points additional coals must be tested to verify this observation. The variation of conversion with total and pyritic sulphur is shown in Figures 7 and 8. Although the figures illustrate that high levels of total and pyritic sulphur may positively affect coal conversion, they also show that other coal properties influence product distribution.

Coal conversion is plotted as a function of carbon content in Figure 9. Conversion to toluene solubles plus gases was found to increase as carbon content decreased from 82 to 69%. The effect of

coal rank on conversion is shown in Figure 10. Coal rank is presented in terms of maximum reflectance of vitrinite (% R_o max.). Figure 10 shows that oil yield increases significantly as rank decreases. The highest oil yield, 57%, was produced from Illinois No. 6 coal, R_o max. 0.62%.

In conclusion, the order of reactivity was found to be: Illinois No. 6 > Prince Hub > Lingan Harbour. The difference in reactivity observed between Illinois No. 6 and the Nova Scotia coals is primarily attributable to rank.

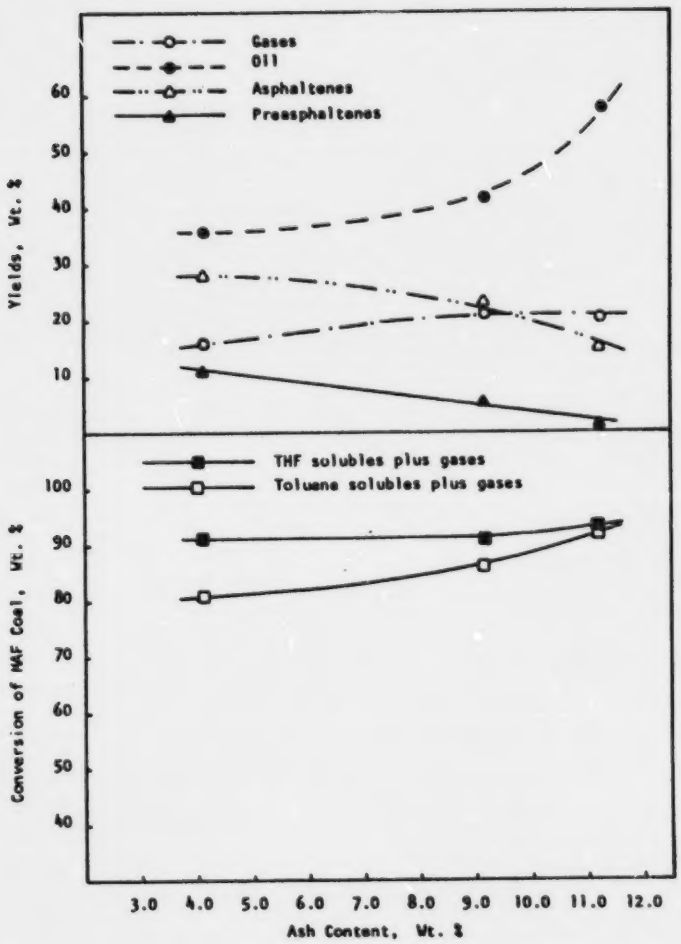


Figure 6 - VARIATION OF COAL CONVERSION WITH ASH CONTENT

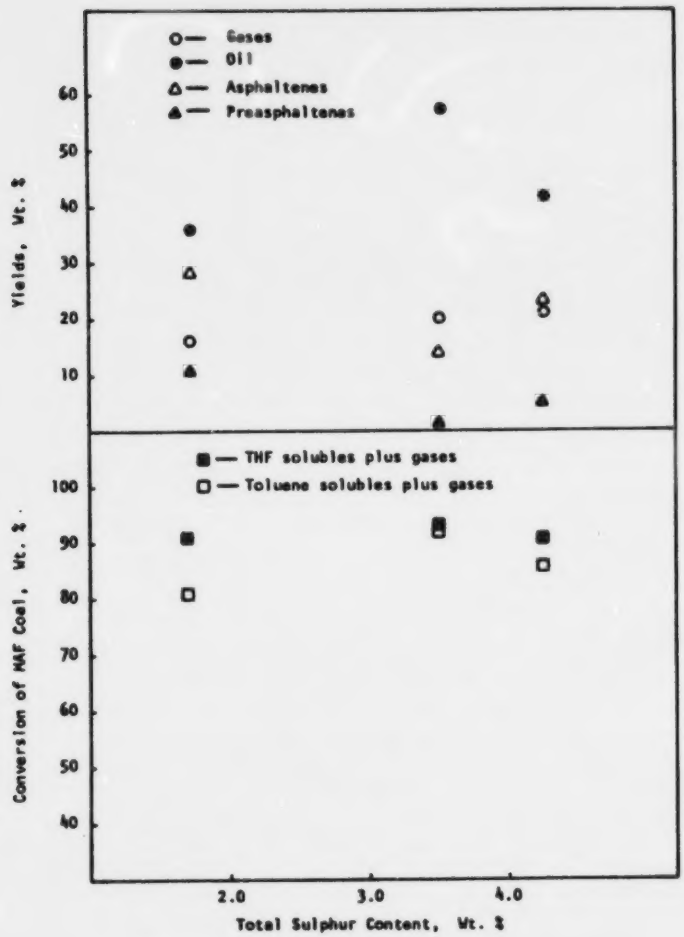


Figure 7 - VARIATION OF COAL CONVERSION WITH TOTAL SULPHUR CONTENT

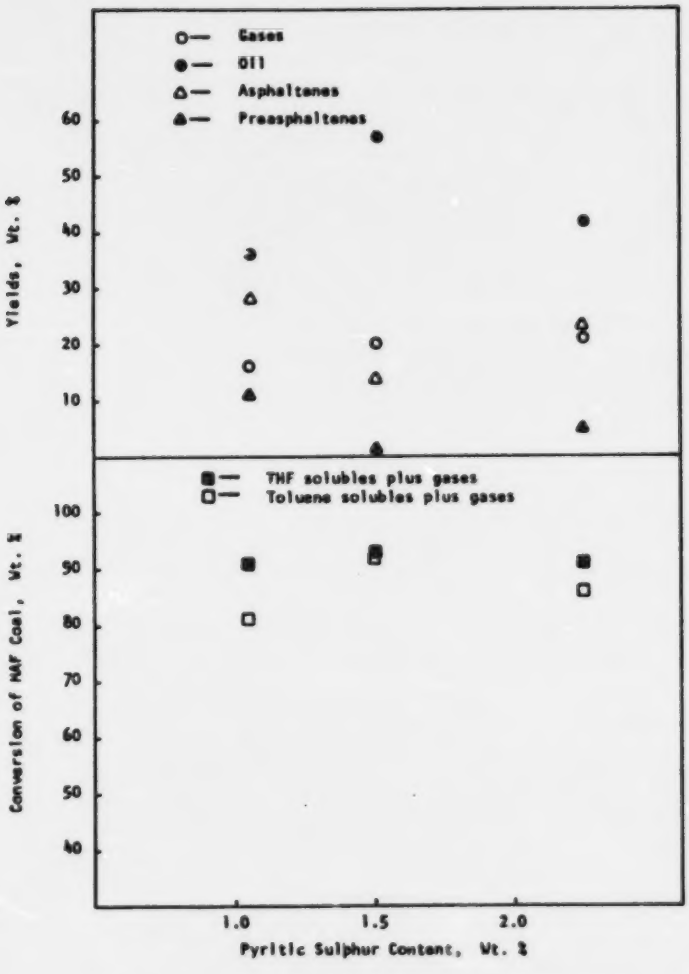


Figure 8 - VARIATION OF COAL CONVERSION WITH PYRITIC SULPHUR CONTENT

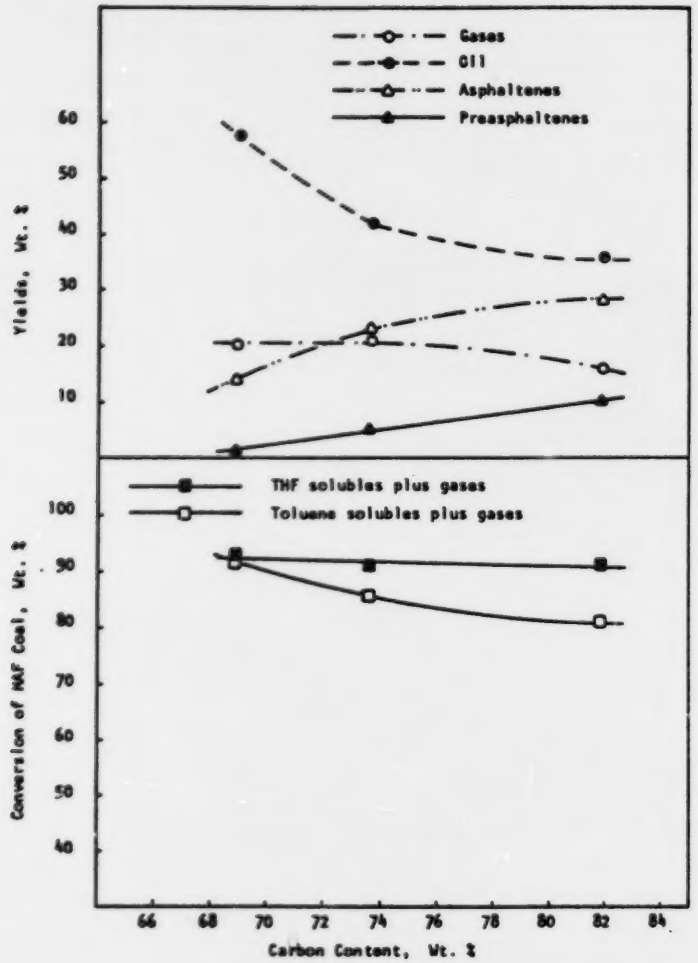


Figure 9 - COAL CONVERSION AS A FUNCTION OF CARBON CONTENT

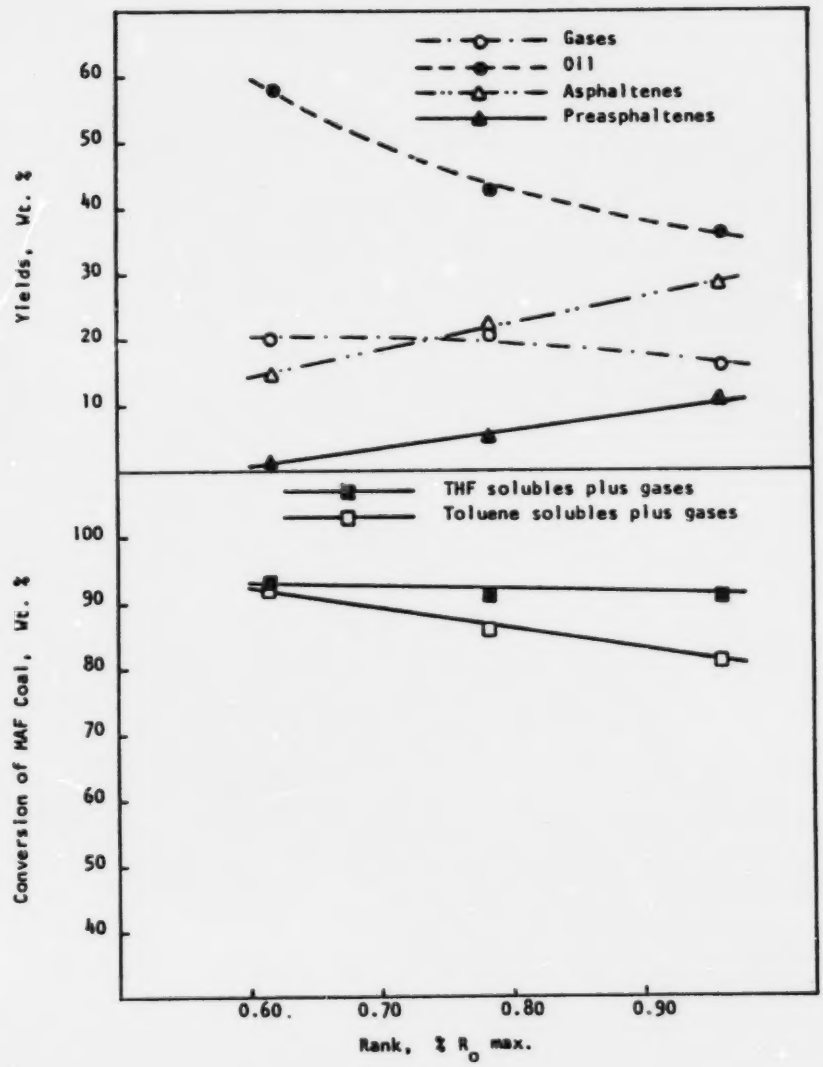


Figure 10 - EFFECT OF RANK ON COAL CONVERSION

6.3 Improved Performance on Lingan Coal at Increased Severity

During Task 6.2 the scope of our test program was revised. According to the original work plan solvent recycle experiments with Lingan coal were scheduled to follow operations with Illinois No. 6. However, because of the low oil yields from Lingan coal the experimental program was revised. The revised program had two main objectives: First, to conduct duplicate runs on Prince coal as described in the previous section. Second, to carry out additional experiments with Lingan coal at increased severity in an attempt to achieve higher oil yields. The operating conditions and test results for the higher severity operations are given in Table 12.

Severity was increased step-wise in advancing from Run Nos. 5021 to 5024. In Run no. 5021 the level of Nalco NM 502 catalyst was increased to 20 Wt. %, other operating conditions were similar to those of Run No. 5011. The effect of the higher catalyst concentration on product distribution was small. Temperature was increased to 460°C in Run No. 5022 and slurry feedrate was reduced to 0.7 kg/hr in Run No. 5023. The changes in temperature and slurry feedrate resulted in a gradual but persistent shift towards lighter products. In Run No. 5024 pressure was increased to 20.68 MPa and 9% H vehicle oil was used. This run produced the highest oil yield, 47.1% and the lowest yield of asphaltene/preasphaltene components, 18.5%. Run No. 5026 was carried out at 460°C, 20.68 MPa and 10 Wt. % catalyst. The reduction in catalyst concentration had a significant negative effect on product distribution.

TABLE 12

CATALYTIC HYDROGENATION OF LINGAN COAL AT INCREASED SEVERITY

RUN NO.	5011	5021	5022	5023	5024	5026
Temperature, °C	451	451	460	460	460	460
Back Pressure, MPa	17.24	17.24	17.24	17.24	20.68	20.68
Slurry Feedrate, kg/hr	0.942	0.940	0.948	0.705	0.698	0.703
H_2 Treatrate, Wt. % Dry Coal	12.8	12.6	12.3	13.0	13.0	13.0
Catalyst Level, Wt. % maf Coal	10	20	20	20	20	10
H_2 Consumption, Wt. % maf Coal	4.0	5.1	5.0	7.2	6.6	6.4
CONVERSIONS, WT. % OF maf COAL						
Pentane Solubles plus Gases	52.5	55.6	61.9	66.7	72.2	65.5
Toluene Solubles plus Gases	80.6	83.4	85.7	86.8	89.1	86.8
THF Solubles plus Gases	91.2	89.5	89.7	90.9	90.7	91.7
PRODUCT DISTRIBUTION, WT. % OF maf COAL						
Gases	16.4	18.1	21.4	24.3	25.1	23.2
Oil	36.1	37.5	40.5	42.4	47.1	42.3
Asphaltenes	28.1	27.8	23.8	20.1	16.9	21.3
Preasphaltenes	10.6	6.1	4.0	4.1	1.6	4.9

Coal concentration in feed slurry; 29.5 Wt. %.
Catalyst used: Nalco NM 502.
7.5% H vehicle oil used in Run Nos. 5021, 5022 and 5023.
9.0% H vehicle oil used in Run Nos. 5024 and 5026.
Detailed product distribution given in Appendix B.

These catalytic hydrogenation runs suggest that oil yields in the range of 50% can be produced from Lingan Harbour and Prince Hub coals in a single-stage catalytic process. Furthermore, the tests suggest that in order to achieve oil yields in this range the Nova Scotia coals would require higher catalyst consumption rates than Illinois No. 6. In fact, it appears that catalyst requirements would be 2 to 3 times higher for the Nova Scotia coals.

TASK 7 - TWO-STAGE THERMAL/CATALYTIC LIQUEFACTION

The purpose of Task 7 was to initiate experimental work on two-stage thermal/catalytic liquefaction. In this process configuration coal is solubilized in the thermal first stage and subsequently upgraded in the catalytic second stage. By using two separate reaction zones the thermal dissolution and the catalytic upgrading steps can be independently optimized to maximize efficiency and control product distribution and quality. It is expected that the two-stage processing scheme will improve hydrogen utilization, increase distillate yield and reduce the production of hydrocarbon gases.

During Phase V testwork was carried out on both stages in a non-integrated mode.

7.1 First Stage Dissolution Studies

A series of eight experiments were conducted according to a factorial design to investigate the effects of temperature, pressure and coal space velocity on conversion in the thermal stage. Lingan Harbour coal was used in these experiments. Hydrogenated anthracene oil containing 9.0% H was used as the donor vehicle. The concentration of coal in the feed slurry was fixed at 30 wt. %. Hydrogen treatrate was ca.7.5 Wt. % based on dry coal. The operating conditions investigated were as follows:

Temperature, ^{o}C	440, 460
Back Pressure, MPa	6.89, 10.34
Slurry Feedrate, kg/hr	1, 2

The objective in the thermal stage is to affect high coal conversion and low hydrocarbon gas production. Results presented in Table 13 show that operating conditions can be selected to achieve the desired objective. A statistical evaluation of the data was carried out to determine the significance of the observed effects. The response variables selected for the statistical analysis included gas make, oil yield, asphaltenes and preasphaltenes. A standard analysis of variance was used to establish confidence levels. Details are presented in Appendix H.

Temperature was found to be highly significant with respect to the production of gases and preasphaltenes. Increasing the temperature from 440 to 460°C produced higher gas yields and lower yields of preasphaltenes. For example, a comparison of Run No. 5031 at 440°C and Run No. 5029 at 460°C shows the following shift in product distribution:

- gases increased from 12.5 to 20.9%
- preasphaltenes decreased from 24.1 to 18.3%.

The statistical evaluation shows that the effect of pressure, over the range 6.89 to 10.34 MPa, is not highly significant with regard to any of the response variables evaluated.

Slurry feedrate was found to significantly affect the yields of gases, asphaltenes and preasphaltenes. Increasing the slurry feedrate produced lower yields of gases and asphaltenes and higher

TABLE 13

SUMMARY OF RESULTS FROM FIRST STAGE DISSOLUTION STUDIES

RUN NO.	5027	5028	5029	5030	5031	5032	5033	5034
Temperature, °C	460	460	460	440	440	460	440	440
Back Pressure, MPa	6.89	10.34	6.89	10.34	6.89	10.34	6.89	10.34
Slurry Feedrate, kg/hr	2.010	1.042	1.005	2.024	1.026	2.008	2.064	1.036
H_2 Consumption, Wt. % maf Coal	1.0	0.9	0.4	0.4	0.4	1.0	0.8	0.9
CONVERSIONS, WT. % OF maf COAL								
Pentane Solubles plus Gases	35.0	38.5	38.7	23.7	32.0	36.0	25.3	32.0
Toluene Solubles plus Gases	60.8	65.2	65.5	50.8	59.3	61.7	50.5	60.3
THF Solubles plus Gases	83.4	84.6	83.8	82.3	83.4	84.8	81.7	84.4
PRODUCT DISTRIBUTION, WT. % OF maf COAL								
Gases	15.0	19.6	20.9	9.2	12.5	14.7	8.9	12.3
Oil	20.0	18.9	17.8	14.5	19.5	21.3	16.4	19.7
Asphaltenes	25.8	26.7	26.8	27.1	27.3	25.7	25.2	28.3
Preasphaltenes	22.6	19.4	18.3	31.5	24.1	23.1	31.2	24.1

Lingan coal used for Run Nos. 5027-5034 inclusive.
Coal concentration in feed slurry; 30 Wt. %.
Detailed product distribution given in Appendix B.

levels of preasphaltenes. For example, a comparison of Run No. 5031 at 1 kg/hr and Run No. 5033 at 2 kg/hr shows the following shift in product distribution:

- gases decreased from 12.5 to 8.9%
- asphaltenes decreased from 27.3 to 25.2%
- preasphaltenes increased from 24.1 to 31.2%.

It is well known that hydrogen deficient coal-derived liquids retrograde on storage. Hence, prior to undertaking the non-integrated two-stage experiments described in Task 7.2 tests were carried out to investigate the storage instability of raw product from the thermal stage. The main purpose of this work was to determine the extent of regressive reactions in raw product stored at controlled conditions for several days.

The tests were conducted using separator bottoms from Run No. 5028. Representative samples of this material were stored under an argon atmosphere at 101.3 kPa and under hydrogen at 6.89 MPa. Both tests were carried out at room temperature since separator bottoms are recovered from the coal liquefaction unit at or near room temperature. The tests were initiated on the day of Run No. 5028 and samples for analysis were taken from the storage vessels after 1, 3, 7 and 14 days. Product stability was monitored by solvent analysis using pentane and toluene.

the storage tests showed that:

(a) Raw product can be held at room temperature under argon for about 3 days without significant changes in the levels of oil and asphaltenes.

(b) After 14 days of storage under argon the quantity of asphaltenes in the raw product was 6.7% higher than the amount originally present.

(c) Levels of oil and asphaltenes in the raw product stored under hydrogen at 6.89 MPa did not change significantly over the 14 day period.

7.2 Non-Integrated Two-Stage Liquefaction Trials

After completion of the first stage dissolution studies bench unit operations were concentrated on non-integrated two-stage thermal/catalytic liquefaction of Lingan Harbour coal. In these experiments thermal stage product was collected over a 10-13 hr operating period and subsequently upgraded. The thermal dissolution and catalytic upgrading steps were separated by approximately 48 hours. A total of three non-integrated trials were carried out during Phase V varying the operating conditions in both stages. A summary of operating conditions and results are shown in Table 14.

In Run No. 5035 the thermal stage was operated at 460°C, 10.34 MPa and a slurry feedrate of 2 kg/hr. The catalytic upgrading step was carried out at 390°C, 20.68 MPa and a slurry feedrate of 1.1 kg/hr. This combination of slurry feedrates provides a total residence

TABLE 14

RESULTS OF NON-INTEGRATED TWO-STAGE LIQUEFACTION TRIALS WITH LINGAN COAL

RUN NO.	5035 I	5035 II	5035 I & II	5036 I	5036 II	5036 I & II	5037 I	5037 II	5037 I & II
Temperature, °C	460	390		460	430		440	430	
Back Pressure, MPa	10.34	20.68		10.34	17.24		17.24	17.24	
Slurry Feedrate, kg/hr	2.073	1.142		2.089	0.765		2.028	0.734	
Catalyst, Wt. % maf Coal	0.0	10		0.0	20		0.0	20	
H_2 Consumption, Wt. % maf Coal	0.6	1.1	1.7	0.6	2.7	3.3	0.9	3.2	4.1
CONVERSIONS, WT. % OF maf COAL									
Pentane Solubles plus Gases	34.8	42.7	42.7	33.8	60.4	60.4	24.1	55.6	55.6
Toluene Solubles plus Gases	62.0	73.7	73.7	61.3	85.7	85.7	52.0	83.5	83.5
THF Solubles plus Gases	84.1	89.5	89.5	84.4	90.4	90.4	84.0	89.8	89.8
PRODUCT DISTRIBUTION, WT. % OF maf COAL									
Gases	14.8	4.5	19.3	13.7	10.1	23.8	9.2	9.5	18.7
Oil	20.0		23.4	20.1		36.6	14.9		36.9
Asphaltenes	27.2		31.0	27.5		25.3	27.9		27.9
Preasphaltenes	22.1		15.8	23.1		4.7	32.0		6.3

Coal concentration in feed slurry; 30 Wt. %.
Nalco NM 502 catalyst used in Runs Nos. 5035 II, 5036 II and 5037 II.
9% H vehicle oil used in Run Nos. 5035 I, 5036 I and 5037 I.
Detailed product distribution given in Appendix B.

time of ca. 60 minutes. Nalco NM-502 catalyst was used at a 10 Wt. % level. Coal conversion was 84.1% in the thermal stage compared with 89.5% in the catalytic. The final product distribution was as follows: 19.3% gases, 23.4% oil, 31.0% asphaltenes and 15.8% preasphaltenes.

In consideration of the low oil yield and the low hydrogen consumption, 1.7%, it was decided to increase severity in the upgrading step. This was accomplished in Run No. 5036 by raising the temperature to 430°C, increasing the catalyst concentration to 20 Wt. % and decreasing the slurry feedrate to 0.8 kg/hr. Run No. 5036 produced 90.4% coal conversion. The product distribution was as follows: 23.8% gases, 36.6% oil, 25.3% asphaltenes and 4.7% preasphaltenes. Upgrading at increased severity produced higher yields of hydrocarbon gas and oil as expected. However, oil yield was still below the target level of +50%.

Several factors influenced the selection of operating conditions for Run No. 5037. First, a limited inventory of 9% H vehicle oil permitted only one further trial. Second, based on our operating experience the thermal stage can be operated to achieve 83-86% coal conversion with significantly less than 14% gas make as in Run Nos. 5035 I and 5036 I. Third, the initial thermal products may, in fact, undergo regressive (condensation) reactions due to thermal cycling between stages. This can render the initial thermal products unresponsive to subsequent catalytic conversion and increase the production of light gas. Finally, on completion of Phase V a second reactor will be added to the existing coal liquefaction unit to permit

integrated two-stage operations. It is planned to operate both stages at the same pressure to eliminate interstage pumping.

In consideration of these factors the thermal stage of Run No. 5037 was operated at 440°C to reduce gas production. Furthermore, both stages were operated at 17.24 MPa to permit a comparison of performance with future integrated two-stage operations. In fact, such a comparison will illustrate the effect of thermal cycling on conversion and product selectivity.

Coal conversion in Run No. 5037 I was similar to that of Run No. 5036 I, 84%. However, reducing the temperature resulted in a lower gas make, 9% as compared with 14%. Catalytic upgrading of the thermal stage effluent produced an oil yield of 36.9%. Although the oil yields in Run No. 5036 and 5037 were essentially the same, 37%, it should be noted that more oil was produced in the upgrading step of Run No. 5037. The results of these experiments suggest that reducing slurry space velocity in the catalytic stage will increase the conversion of asphaltenes and preasphaltenes to oil with minimum gas production.

In the thermal dissolution step of this two-stage liquefaction process the conversion of coal is highly dependent on the stabilization of pyrolytically formed free-radicals. These free-radicals are normally stabilized by accepting hydrogen from donor molecules. Thus, in the first stage a structural requirement of the vehicle oil is that it contains transferable hydrogen.

The elemental analysis presented in Appendix D shows higher levels of hydrogen in the pentane, toluene and THF soluble fractions from Run No. 5037 I than in similar fractions from Run Nos. 5028 and 5036 I. This data in combination with the structural analysis (proton distribution) of 9% H vehicle oil suggest that at the medium severity operating conditions of Run No. 5037 I further free-radical formation is possible and/or that stabilization of free radicals may still be in progress. Therefore the first-stage effluent should be responsive to catalytic upgrading in a closely coupled two-stage system.

The analysis of distillate products from separator bottoms of selected runs is given in Appendix E. Elemental analysis shows that the hydrogen content of the distillate product from Run No. 5037 II was ca. 9%. Furthermore, H-NMR analysis illustrate that transferrable hydrogen, H_{β}, was maintained at a high level. These results suggest that a recycle oil with high donor capacity can be produced from Lingan Harbour coal. This is essential to the successful operation of two-stage thermal/catalytic liquefaction.

CONCLUSIONS AND RECOMMENDATIONS

Phase V of our experimental studies on the direct hydroliquefaction of Nova Scotia coals has been completed. This phase of work was primarily concentrated on single-stage catalytic hydrogenation in a bench-scale continuous-flow unit. In addition, testwork was initiated on two-stage thermal/catalytic liquefaction and efforts were made to compare the reactivity of Nova Scotia coals with that of Illinois No. 6. The latter being a highly reactive American coal.

A total of thirty-eight continuous experiments were conducted with Harbour and Hub seam coals from the Sydney Coalfield and Illinois No. 6 from the Burning Star Mine. The runs were made in a once-through mode using hydrogenated anthracene oil as vehicle.

Duplicate runs were carried out with Lingan Harbour and Illinois No. 6 coals under thermal liquefaction conditions. Hydrogenated anthracene oil, 9% H content, was used as donor solvent. The experiments with Illinois No. 6 produced 93% coal conversion compared to 86% with Lingan Harbour. Oil yields were 30 and 22% respectively. These results show that Illinois No. 6 coal is more reactive than Lingan Harbour at thermal liquefaction conditions.

Catalytic hydrogenation studies were focused primarily on Lingan Harbour coal. Two commercially available hydrotreating catalysts, Nalco NM 502 and American Cyanamid HDS-1442, were

evaluated. The former is an alumina-supported $NiO-MoO_3$ catalyst, the latter is an alumina-supported $CoO-MoO_3$ catalyst. In this study pulverized catalyst was added to the feed slurry to simulate single-stage catalytic liquefaction.

On completion of baseline experiments a series of five runs was made according to an optimization technique known as Simplex Self-Directing Evolutionary Operation (SSEVOP). The runs were made varying temperature (451-467°C), catalyst level (7 - 10 Wt. %), hydrogen treatrate (11 - 15 Wt. %) and slurry feedrate (0.92 - 1.32 kg/hr.). Hydrogenated anthracene oil, 7.5% H, was used as vehicle. Pressure was fixed at 17.24 MPa and Nalco NM 502 was used as catalyst. It was found that although coal conversions were high, 90%, oil yields did not exceed 37%. Operations with the Co/Mo catalyst demonstrated that it was less effective in terms of coal hydrogenation activity than the Ni/Mc catalyst.

Additional experiments were conducted with Lingan Harbour coal at increased severity. It was found that operating conditions could be varied to exercise considerable control over product distribution. The highest oil yield, 47%, was achieved at 460°C, 20.68 MPa, 20 Wt. % catalyst and a slurry feedrate of 0.70 kg/hr.

A difference in product yield was observed in single-stage catalytic operations with Lingan Harbour, Prince Hub and Illinois No. 6 coals. Although coal conversions were quite similar, 90.5 to

93.6%, product distributions varied widely. Duplicate experiments with Illinois No. 6 coal produced 57% oil compared to 42% with Prince Hub and 36% with Lingan Harbour. The three coals studied differ in both their chemical and petrographic analysis. However, the difference in reactivity observed between the Nova Scotia coals and Illinois No. 6 is primarily attributed to rank.

The test program carried out on catalytic hydrogenation suggests that oil yields in the range of 50% can be produced from Lingan Harbour and Prince Hub seam coals in a conventional single-stage catalytic process. However, the experiments show that in order to achieve oil yields in this range the Nova Scotia coals would require higher catalyst consumption rates than Illinois No. 6. In fact, it appears that catalyst consumption rates would be 2 to 3 times higher for the Nova Scotia coals.

On completion of the catalytic hydrogenation experiments work began on two-stage thermal/catalytic coal liquefaction. In the two-stage process under investigation coal is solubilized in the thermal first stage and subsequently upgraded in the catalytic second stage. By using this approach the dissolution step and the upgrading step can be separately optimized to maximize efficiency and control product distribution and quality. It is expected that the two-stage process configuration will improve hydrogen utilization, increase distillate yield and reduce the production of hydrocarbon gases. During Phase V testwork was carried out on both stages in a non-integrated mode.

A 2^3 factorial experiment was conducted to assess first-stage operating conditions. As a result of a statistical evaluation of the test results it was found that:

(a) Temperature, over the range 440 to 460°C, affects the conversion of coal to gases and preasphaltenes. In general, higher yields of gases and lower yields of preasphaltenes were observed at 460°C.

(b) Pressure, over the range 6.89 to 10.34 MPa had little effect on overall conversion.

(c) Slurry feedrate, over the range 1 to 2 kg/hr affected the yields of gases, asphaltenes and preasphaltenes. Increasing the feedrate produced lower yields of gases and asphaltenes and higher yields of preasphaltenes.

Following the thermal dissolution studies three non-integrated trials were carried out using Nalco NM 502 in the catalytic stage. In these experiments thermal stage product was collected and subsequently upgraded. The thermal dissolution and catalytic upgrading stages were separated by approximately 48 hours. High coal conversion, 90%, was achieved in these trials. Oil yields ranged from 23 - 37%. The results of these experiments suggest that reducing slurry space velocity in the catalytic stage will increase oil yield with minimum gas production.

It was not intended to pursue this process in a non-integrated mode, because yields may be influenced by the

method of sequencing of the stages. The initial thermal products can undergo regressive (condensation) reactions which render them unresponsive to subsequent catalytic conversion and which increase the light gas yield. This can occur during first-stage dissolution, if there is limited donor capacity for free-radical stabilization, and upon storage and/or thermal cycling between stages. The latter effects can be avoided by operating the two stages in direct sequence. Test results suggest that first-stage product will be highly responsive to catalytic upgrading in a closely coupled two-stage system. Furthermore, data generated suggest that a recycle oil with high donor capacity can be produced from Lingan Harbour coal.

Based on our findings we recommend that experimental work be undertaken on integrated two-stage thermal/catalytic liquefaction as planned. In addition, it is recommended that following further scoping of first and second-stage operating conditions a series of continuous runs be made in a sequential format designed to generate an equilibrium or steady-state recycle oil from a Nova Scotia coal.

At present, conventional coal liquefaction is not commercially viable in Eastern Canada. Process developments are required to improve distillate yield and/or reduce plant capital costs. Two-stage direct hydroliquefaction offers the potential to enhance distillate yield and improve hydrogen utilization. Extensive testwork is required on two-stage process configurations to determine the extent to which these improvements can be realized.

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APPENDIX A

CONVERSION CALCULATIONS

APPENDIX A

CONVERSION CALCULATIONS FOR CONTINUOUS COAL LIQUEFACTION EXPERIMENTS

Calculations are based on operations during the steady state sampling period.

CONVERSIONS, WT. %:

$$\text{Pentane sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic pentane insol.})}{\text{maf coal charged}} \times 100$$

$$\text{Toluene sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic toluene insol.})}{\text{maf coal charged}} \times 100$$

$$\text{THF sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic THF insol.})}{\text{maf coal charged}} \times 100$$

$$\text{Gases} = \frac{\text{product gases}}{\text{maf coal charged}} \times 100$$

DEFINITION OF PRODUCTS

Oil = pentane solubles

Asphaltenes = toluene solubles - pentane solubles

Preasphaltenes = THF solubles - toluene solubles

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR CONTINUOUS EXPERIMENTS

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR DONOR SOLVENT LIQUEFACTION EXPERIMENTS

RUN NO.	5001	5002	5003	5004
COAL TESTED	ILLINOIS NO. 6	ILLINOIS NO. 6	LINGAN L	LINGAN L
A. OPERATING CONDITIONS				
Unit Back Pressure, MPa	20.68	20.68	20.68	20.68
Temperatures, °C:				
Reactor (Internal)	460	460	460	460
Slurry Preheater	184	186	173	171
Hot Separator	253	252	254	254
Feedrates:				
Slurry, kg/hr	2.048	2.015	2.059	2.003
Coal, kg/hr	0.614	0.605	0.618	0.601
Hydrogen, L/hr	492.	462.	462.	495.
Hydrogen Treatrate, Wt. % Dry Coal	7.5	7.0	6.8	7.5
Hours on Stream	6.00	6.00	6.00	6.00
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	47.6	47.3	33.5	35.0
Toluene Solubles plus Gases	75.0	77.3	62.7	64.1
Benzene Solubles plus Gases	74.7	75.2	62.1	63.3
THF Solubles plus Gases	92.6	93.1	86.0	86.3
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	2.6	2.9	2.9	3.0
C_2H_6	2.9	2.8	2.4	2.4
C_3H_8	2.7	2.8	2.4	2.4
C_4H_{10}	2.4	2.4	2.2	2.2
CO	0.5	0.6	0.2	0.2
H_2S	1.5	1.6	0.7	0.7
H_2O	5.2	5.5	1.2	2.0
Oil	29.8	28.7	21.5	22.1
Asphaltenes	27.4	30.0	29.2	29.1
Preasphaltenes	17.6	15.8	23.3	22.2
Unreacted Coal	7.4	6.9	14.0	13.7
C. H_2 CONSUMPTION, WT. % OF maf COAL	1.0	0.7	0.9	1.3

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.		5005	5006	5007	5009
COAL TESTED		LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED		NM 502	NM 502	NM 502	NM 502
A.	OPERATING CONDITIONS				
	Unit Back Pressure, MPa	17.24	17.24	17.24	17.24
	Temperatures, °C:				
	Reactor (Internal)	460	460	460	451
	Slurry Preheater	221	219	215	215
	Hot Separator	250	250	251	251
	Feedrates:				
	Slurry, kg/hr	1.054	1.009	0.793	0.925
	Coal, kg/hr	0.315	0.296	0.233	0.273
	Hydrogen, L/hr	246.	258.	384.	355.
	Hydrogen Treatrate, Wt. % Dry Coal	7.5	8.0	15.1	11.0
	Catalyst Level, Wt. % maf Coal	4	8	8	7
	Hours on Stream	6.50	7.10	6.57	6.29
B.	CONVERSIONS OF maf COAL, WT. %				
	Pentane Solubles plus Gases	45.0	50.2	60.1	50.7
	Toluene Solubles plus Gases	76.8	79.8	84.6	79.7
	THF Solubles plus Gases	91.0	90.9	91.6	91.4
C.	PRODUCT YIELDS ON maf COAL, WT. %				
	CH_4	3.7	4.0	5.0	3.4
	C_2H_6	3.2	3.7	4.8	3.1
	C_3H_8	2.8	3.2	3.8	2.6
	C_4H_{10}	2.4	2.6	3.3	2.4
	CO	0.2	0.2	0.2	0.2
	H_2S	0.5	0.5	0.4	0.4
	H_2O	2.3	2.9	4.4	2.1
	Oil	29.9	33.1	38.2	36.5
	Asphaltenes	31.8	29.6	24.5	29.0
	Preasphaltenes	14.2	11.1	7.0	11.7
	Unreacted Coal	9.0	9.1	8.4	8.6
D.	H_2 CONSUMPTION, WT. % OF maf COAL	4.0	4.3	5.6	4.1

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5010	5011	5012	5014
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NM 502	NM 502	NM 502	NM 502
A. OPERATING CONDITIONS				
Unit Back Pressure, MPa	17.24	17.24	17.24	17.24
Temperatures, °C:				
Reactor (Internal)	451	451	467	455
Slurry Preheater	213	213	217	201
Hot Separator	251	252	252	253
Feedrates:				
Slurry, kg/hr	0.925	0.942	0.927	1.319
Coal, kg/hr	0.273	0.278	0.273	0.389
Hydrogen, L/hr	444.	390.	390.	558.
Hydrogen Treatrate, Wt. % Dry Coal	14.9	12.8	13.0	13.0
Catalyst Level, Wt.% maf Coal	7	10	8	8
Hours on Stream	6.02	6.03	6.02	6.00
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	51.2	52.5	59.7	49.0
Toluene Solubles plus Gases	79.5	80.6	84.9	77.0
THF Solubles plus Gases	91.6	91.2	92.1	90.5
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	3.5	3.5	5.6	3.3
C_2H_6	3.2	3.3	5.4	3.1
C_3H_8	2.6	2.5	4.5	2.4
C_4H_{10}	2.6	2.7	3.8	2.7
CO	0.2	0.1	0.5	0.3
H_2S	0.4	0.3	0.2	0.4
H_2O	4.0	4.0	5.1	3.7
Oil	34.7	36.1	34.6	33.1
Asphaltenes	28.3	28.1	25.2	28.0
Preasphaltenes	12.1	10.6	7.2	13.5
Unreacted Coal	8.4	8.8	7.9	9.5
D. H_2 CONSUMPTION, WT. % OF maf COAL	4.8	4.0	4.8	4.3

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5015	5016	5017	5018
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	ILLINOIS NO. 6
CATALYST TESTED	NM 502	HDS 1442	NM 502	NM 502
A. OPERATING CONDITIONS				
Unit Back Pressure, MPa	17.24	17.24	17.24	17.24
Temperatures, °C:				
Reactor (Internal)	455	460	460	451
Slurry Preheater	216	219	220	220
Hot Separator	251	251	251	252
Feedrates:				
Slurry, kg/hr	0.926	0.858	0.882	0.936
Coal, kg/hr	0.273	0.253	0.260	0.276
Hydrogen, L/hr	360.	390.	348.	390.
Hydrogen Treatrate, Wt. % Dry Coal	12.0	14.0	12.2	13.1
Catalyst Level, Wt. % maf Coal	8	8	8	10
Hours on Stream	5.97	6.04	6.07	6.00
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	52.6	55.9	60.9	78.0
Toluene Solubles plus Gases	80.7	81.6	83.8	91.9
THF Solubles plus Gases	90.7	90.9	91.9	93.6
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	3.8	4.7	4.5	2.7
C_2H_6	3.6	4.7	4.9	3.0
C_3H_8	2.9	3.6	4.3	2.8
C_4H_{10}	3.0	3.6	3.7	2.7
CO	0.4	0.3	0.2	0.2
H_2S	0.3	0.3	0.2	1.8
H_2O	4.3	4.8	4.9	* 7.1
Oil	34.3	33.9	38.2	57.7
Asphaltenes	28.1	25.7	22.9	13.9
Preasphaltenes	10.0	9.3	8.1	1.7
Unreacted Coal	9.3	9.1	8.1	6.4
D. H_2 CONSUMPTION, WT. % OF maf COAL	4.1	4.4	3.7	5.9

* - Water calculated based on water production for entire run.

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5019	5020	5020A	5021
COAL TESTED	ILLINOIS NO. 6	PRINCE D	PRINCE D	LINGAN L
CATALYST TESTED	NM 502	NM 502	NM 502	NM 502
A. OPERATING CONDITIONS				
Unit Back Pressure, MPa	17.24	17.24	17.24	17.24
Temperatures, °C:				
Reactor (Internal)	451	451	451	451
Slurry Preheater	220	216	220	215
Hot Separator	252	251	300	299
Feedrates:				
Slurry, kg/hr	0.953	0.922	0.915	0.940
Coal, kg/hr	0.281	0.272	0.270	0.277
Hydrogen, L/hr	390.	390.	384.	384.
Hydrogen Treatrate, Wt. % Dry Coal	13.0	13.2	13.0	12.6
Catalyst Level, Wt. % maf Coal	10	10	10	20
Hours on Stream	6.05	6.08	6.00	8.00
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	76.9	63.4	61.8	55.6
Toluene Solubles plus Gases	91.7	85.6	85.9	83.4
THF Solubles plus Gases	93.3	90.6	90.5	89.5
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	2.8	3.4	3.1	3.3
C_2H_6	2.8	3.7	3.3	3.2
C_3H_8	2.9	2.9	2.5	2.4
C_4H_{10}	2.7	2.9	3.1	3.4
CO	0.3	0.3	0.3	0.1
H_2S	1.7	1.3	1.3	0.0
H_2O	* 6.9	* 6.4	7.2	5.7
Oil	56.8	42.5	41.0	37.5
Asphaltenes	14.8	22.2	24.1	27.8
Preasphaltenes	1.6	5.0	4.6	6.1
Unreacted Coal	6.7	9.4	9.5	10.5
D. H_2 CONSUMPTION, WT. % OF maf COAL	5.7	4.6	5.1	5.1

* - Water calculated based on water production for entire run.

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5022	5023	5024	5026
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NM 502	NM 502	NM 502	NM 502
A. OPERATING CONDITIONS				
Unit Back Pressure, MPa	17.24	17.24	20.68	20.68
Temperatures, °C:				
Reactor (Internal)	460	460	460	460
Slurry Preheater	215	223	225	227
Hot Separator	299	302	304	303
Feedrates:				
Slurry, kg/hr	0.948	0.705	0.698	0.703
Coal, kg/hr	0.280	0.208	0.224	0.207
Hydrogen, L/hr	378.	294.	294.	294.
Hydrogen Treatrate, Wt. % Dry Coal	12.3	13.0	13.0	13.0
Catalyst Level, Wt. % maf Coal	20	20	20	10
Hours on Stream	8.03	6.50	6.49	6.50
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	61.9	66.7	72.2	65.5
Toluene Solubles plus Gases	85.7	86.8	89.1	86.8
THF Solubles plus Gases	89.7	90.9	90.7	91.7
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	4.1	5.0	4.3	4.4
C_2H_6	4.9	5.1	4.9	5.0
C_3H_8	3.0	3.9	4.2	4.1
C_4H_{10}	3.2	3.4	4.2	4.3
CO	0.1	0.1	0.1	0.2
H_2S	0.0	0.0	0.0	0.2
H_2O	6.1	6.6	7.2	4.7
NH_3	0.0	0.2	0.2	0.3
Oil	40.5	42.4	47.1	42.3
Asphaltenes	23.8	20.1	16.9	21.3
Preasphaltenes	4.0	4.1	1.6	4.9
Unreacted Coal	10.3	9.1	9.3	8.3
D. H_2 CONSUMPTION, WT. % OF maf COAL	5.0	7.2	6.6	6.4

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR FIRST STAGE DISSOLUTION EXPERIMENTS

RUN NO.	5027	5028	5029	5030
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NONE	NONE	NONE	NONE
A. OPERATING CONDITIONS				
Unit Back Pressure, MPa	6.89	10.34	6.89	10.34
Temperatures, °C:				
Reactor (Internal)	460	460	460	440
Slurry Preheater	196	220	222	177
Hot Separator	306	300	300	303
Feedrates:				
Slurry, kg/hr	2.010	1.042	1.005	2.024
Coal, kg/hr	0.603	0.313	0.302	0.607
Hydrogen, L/hr	498.	252.	210.	504.
Hydrogen Treatrate, Wt.% Dry Coal	7.5	7.4	6.3	7.6
Catalyst Level, Wt. % maf Coal	0.0	0.0	0.0	0.0
Hours on Stream	6.09	6.03	6.05	6.03
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	35.0	38.5	38.7	23.7
Toluene Solubles plus Gases	60.8	65.2	65.5	50.8
THF Solubles plus Gases	83.4	84.6	83.8	82.3
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	3.3	4.9	5.3	1.6
C_2H_6	3.0	4.3	4.6	1.7
C_3H_8	2.2	3.5	3.8	1.1
C_4H10	2.7	3.2	3.1	2.1
CO	0.3	0.4	0.4	0.2
H_2S	0.8	0.7	0.8	0.8
H_2O	2.6	2.6	2.9	1.7
NH_3	0.1	0.0	0.0	0.0
Oil	20.0	18.9	17.8	14.5
Asphaltenes	25.8	26.7	26.8	27.1
Preasphaltenes	22.6	19.4	18.3	31.5
Unreacted Coal	16.6	15.4	16.2	17.7
D. H_2 CONSUMPTION, WT. % OF maf COAL	1.0	0.9	0.4	0.4

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR FIRST STAGE DISSOLUTION EXPERIMENTS

RUN NO.	5031	5032	5033	5034
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NONE	NONE	NONE	NONE
A. OPERATING CONDITIONS				
Unit Back Pressure, MPa	6.89	10.34	6.89	10.34
Temperatures, °C:				
Reactor (Internal)	440	460	440	440
Slurry Preheater	223	185	188	238
Hot Separator	301	304	307	302
Feedrates:				
Slurry, kg/hr	1.026	2.008	2.064	1.036
Coal, kg/hr	0.308	0.602	0.619	0.311
Hydrogen, L/hr	252.	504.	504.	300.
Hydrogen Treatrate, Wt. % Dry Coal	7.5	7.5	7.5	8.8
Catalyst Level, Wt. % maf Coal	0.0	0.0	0.0	0.0
Hours on Stream	6.00	7.03	6.00	6.07
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	32.0	36.0	25.3	32.0
Toluene Solubles plus Gases	59.3	61.7	50.5	60.3
THF Solubles plus Gases	83.4	84.8	81.7	84.4
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	2.7	3.3	1.7	2.7
C_2H_6	2.3	2.8	1.8	2.3
C_3H_8	1.9	2.4	0.9	1.6
C_4H_{10}	2.4	2.7	1.7	2.4
CO	0.3	0.4	0.2	0.2
H_2S	0.4	0.8	0.6	0.8
H_2O	2.5	2.3	2.0	2.3
NH_3	0.0	0.0	0.0	0.0
Oil	19.5	21.3	16.4	19.7
Asphaltenes	27.3	25.7	25.2	28.3
Preasphaltenes	24.1	23.1	31.2	24.1
Unreacted Coal	16.6	15.2	18.3	15.6
D. H_2 CONSUMPTION, WT. % OF maf COAL	0.4	1.0	0.8	0.9

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR NON-INTEGRATED TWO-STAGE LIQUEFACTION TESTS

RUN NO.	5035 I	5035 II	5035 I/II
COAL TESTED	LINGAN L	LINGAN L	
CATALYST TESTED		NM 502	
A. OPERATING CONDITIONS			
Unit Back Pressure, MPa	10.34	20.68	
Temperatures, °C:			
Reactor (Internal)	460	390	
Slurry Preheater	186	215	
Hot Separator	306	303	
Feedrates:			
Slurry, kg/hr	2.073	1.142	
Coal, kg/hr	0.622		
Hydrogen, L/hr	504.	480.	
Hydrogen Treatrate, Wt. % Dry Coal	7.4	13.1	
Catalyst Level, Wt. % maf Coal	0.0	10	
Hours on Stream	13.00	6.17	
B. CONVERSIONS OF maf COAL, WT. %			
Pentane Solubles plus Gases	34.8	42.7	42.7
Toluene Solubles plus Gases	62.0	73.7	73.7
THF Solubles plus Gases	84.1	89.5	89.5
C. PRODUCT YIELDS ON maf COAL, WT. %			
CH_4	3.2	0.2	3.4
C_2H_6	2.9	1.1	4.0
C_3H_8	2.3	0.1	2.4
C_4H_{10}	2.9	2.5	5.4
CO	0.3	0.0	0.3
H_2S	0.8	0.0	0.8
H_2O	2.4	0.6	3.0
NH_3	0.0	0.0	0.0
Oil	20.0		23.4
Asphaltenes	27.2		31.0
Preasphaltenes	22.1		15.8
Unreacted Coal	15.9	10.5	10.5
D. H_2 CONSUMPTION, WT. % OF maf COAL	0.6	1.1	1.7

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR NON-INTEGRATED TWO-STAGE LIQUEFACTION TESTS

RUN NO. COAL TESTED CATALYST TESTED	5036 I LINGAN L	5036 II LINGAN L NM 502	5036 I/II
A. OPERATING CONDITIONS			
Unit Back Pressure, MPa	~10.34	17.24	
Temperatures, °C:			
Reactor (Internal)	460	430	
Slurry Preheater	188	235	
Hot Separator	305	307	
Feedrates:			
Slurry, kg/hr	2.089	0.765	
Coal, kg/hr	0.627		
Hydrogen, L/hr	504.	360.	
Hydrogen Treatrate, Wt. % Dry Coal	7.3	15.1	
Catalyst Level, Wt. % maf Coal	0.0	20	
Hours on Stream	12.25	7.00	
B. CONVERSIONS OF maf COAL, WT. %			
Pentane Solubles plus Gases	33.8	60.4	60.4
Toluene Solubles plus Gases	61.3	85.7	85.7
THF Solubles plus Gases	84.4	90.4	90.4
C. PRODUCT YIELDS ON maf COAL, WT. %			
CH_4	2.9	1.3	4.2
C_2H_6	2.4	1.7	4.1
C_3H_8	2.0	0.9	2.9
C_4H_{10}	2.6	2.8	5.4
CO	0.3	0.2	0.5
H_2S	0.7	0.0	0.7
H_2O	2.8	3.1	5.9
NH_3	0.0	0.1	0.1
Oil	20.1		36.6
Asphaltenes	27.5		25.3
Preasphaltenes	23.1		4.7
Unreacted Coal	15.6	9.6	9.6
D. H_2 CONSUMPTION, WT. % OF maf COAL	0.6	2.7	3.3

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR NON-INTEGRATED TWO-STAGE LIQUEFACTION TESTS

RUN NO.	5037 I	5037 II	5037 I/5037 II
COAL TESTED	LINGAN L	LINGAN L	
CATALYST TESTED		NM 502	
A. OPERATING CONDITIONS			
Unit Back Pressure, MPa	17.24	17.24	
Temperatures, °C:			
Reactor (Internal)	440	430	
Slurry Preheater	188	232	
Hot Separator	301	306	
Feedrates:			
Slurry, kg/hr	2.028	0.734	
Coal, kg/hr	0.608		
Hydrogen, L/hr	504.	336.	
Hydrogen Treatrate, Wt.% Dry Coal	7.5	15.0	
Catalyst Level, Wt. % maf Coal	0.0	20.	
Hours on Stream	9.53	6.50	
B. CONVERSIONS OF maf COAL, WT. %			
Pentane Solubles plus Gases	24.1	55.6	55.6
Toluene Solubles plus Gases	52.0	83.5	83.5
THF Solubles plus Gases	84.0	89.8	89.8
C. PRODUCT YIELDS ON maf COAL, WT. %			
CH_4	1.6	1.4	3.0
C_2H_6	1.5	1.8	3.3
C_3H_8	1.0	0.9	1.9
C_4H_{10}	2.4	2.9	5.3
CO	0.2	0.1	0.3
H_2S	0.6	0.0	0.6
H_2O	1.9	2.4	4.3
NH_3	0.0	0.0	0.0
Oil	14.9		36.9
Asphaltenes	27.9		27.9
Preasphaltenes	32.0		6.3
Unreacted Coal	16.0	10.2	10.2
D. H_2 CONSUMPTION, WT. % OF maf COAL	0.9	3.2	4.1

APPENDIX C

MATERIAL BALANCES FOR CONTINUOUS EXPERIMENTS

APPENDIX C

MATERIAL BALANCES FOR DONOR SOLVENT LIQUEFACTION EXPERIMENTS

RUN NO.	S001	S002	S003	S004
COAL TESTED	ILLINOIS NO. 6	ILLINOIS NO. 6	LINGAN L	LINGAN L
MOISTURE, WT. %	5.17	5.17	1.25	1.25
ASH, WT. % DRY BASIS	11.13	11.13	4.16	4.16
HOURS ON STREAM	6.00	6.00	6.00	6.00
DURATION OF BALANCE PERIOD, HRS	1.00	1.00	1.00	1.00
INPUT (g):				
H_2O	19.5	19.2	7.9	7.7
Ash	66.2	65.1	25.0	24.3
maf Coal	529.	520.	585.	569.
Solvent	1434.	1411.	1441.	1402.
H_2	44.3	41.6	41.6	44.6
Total	2093.	2057.	2101.	2048.
OUTPUT (g):				
H_2O	47.2	47.7	14.7	19.3
Ash	66.7	63.6	25.7	24.8
Organic THF Insolubles[1]	39.3	36.1	82.1	77.8
THF Solubles[2]	1839.	1803.	1864.	1818.
H_2	39.1	37.8	36.5	37.6
Other Gases	66.6	68.3	63.2	62.0
Total	2098.	2057.	2086.	2040.
TOTAL MATERIAL RECOVERY, WT. %	100.2	100.0	99.3	99.6

1 - Unreacted coal

2 - Oil, asphaltenes and preasphaltenes

APPENDIX C

MATERIAL BALANCES FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5005	5006	5007	5009
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NM 502	NM 502	NM 502	NM 502
CATALYST, WT. % maf COAL	4	8	8	7
MOISTURE, WT. %	1.28	1.28	1.28	1.28
ASH, WT. % DRY BASIS	4.10	4.10	4.10	4.10
HOURS ON STREAM	6.50	7.10	6.57	6.29
DURATION OF BALANCE PERIOD, HRS.	2.03	2.35	2.48	2.02
INPUT (g):				
H_2O	8.1	8.9	7.4	7.0
Ash	25.7	28.1	23.4	22.3
maf Coal	602.	658.	547.	521.
Solvent	1484.	1623.	1348.	1278.
H_2	45.0	54.6	85.8	59.9
Catalyst	24.2	52.6	43.7	36.5
Total	2189.	2425.	2055.	1925.
OUTPUT (g):				
H_2O	22.1	28.2	31.2	15.0
*Ash	48.8	76.8	58.4	53.4
Organic THF Insolubles[1]	54.3	59.7	45.9	45.0
THF Solubles[2]	1944.	2150.	1740.	1687.
H_2	20.7	26.1	55.1	38.4
Other Gases	77.2	93.2	95.9	62.7
Total	2167.	2434.	2027.	1902.
TOTAL MATERIAL RECOVERY, WT. %	99.0	100.4	98.6	98.8

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes
* - Catalyst included

APPENDIX C

MATERIAL BALANCES FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5010	5011	5012	5014
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NM 502	NM 502	NM 502	NM 502
CATALYST, WT. % maf COAL	7	10	8	8
MOISTURE, WT. %	1.28	1.28	1.28	1.28
ASH, WT. % DRY BASIS	4.10	4.10	4.10	4.10
HOURS ON STREAM	6.02	6.03	6.02	6.00
DURATION OF BALANCE PERIOD, HRS.	2.02	2.02	2.00	1.43
INPUT (g):				
H_2O	7.0	7.2	7.0	7.1
Ash	22.3	22.7	22.1	22.4
maf Coal	521.	530.	518.	525.
Solvent	1278.	1286.	1266.	1283.
H_2	80.6	70.8	70.2	71.6
Catalyst	36.6	53.0	41.3	41.9
Total	1946.	1970.	1925.	1951.
OUTPUT (g):				
H_2O	27.8	28.4	33.3	26.5
*Ash	53.5	68.5	54.6	62.2
Organic THF Insolubles[1]	43.7	46.4	40.9	50.1
THF Solubles[2]	1696.	1693.	1639.	1698.
H_2	55.8	49.8	45.2	49.1
Other Gases	65.1	65.9	103.6	64.1
Total	1942.	1952.	1917.	1950.
TOTAL MATERIAL RECOVERY, WT. %	99.8	99.1	99.6	99.9

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes
* - Catalyst included.

APPENDIX C

MATERIAL BALANCES FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5015	5016	5017	5018
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	ILLINOIS NO. 6
CATALYST TESTED	NM 502	HDS 1442	NM 502	NM 502
CATALYST, WT. % maf COAL	8	8	8	10
MOISTURE, WT. %	1.28	1.28	1.28	3.17
ASH, WT. % DRY BASIS	4.10	4.10	4.10	11.13
HOURS ON STREAM	5.97	6.04	6.07	6.00
DURATION OF BALANCE PERIOD, HRS.	1.97	2.15	2.02	1.95
INPUT (g):				
H_2O	6.9	7.0	6.7	17.1
Ash	21.8	22.0	21.2	58.0
maf Coal	509.	515.	497.	463.
Solvent	1244.	1260.	1215.	1240.
H_2	63.7	75.5	63.2	68.4
Catalyst	40.6	41.1	39.7	46.4
Total	1886.	1921.	1843.	1893.
OUTPUT (g):				
H_2O	28.9	31.6	31.2	65.0
*Ash	58.2	57.1	52.1	99.8
Organic THF Insolubles[1]	47.4	47.0	40.5	29.8
THF Solubles[2]	1633.	1637.	1590.	1603.
H_2	42.7	52.9	45.0	41.0
Other Gases	71.2	88.6	88.5	60.7
Total	1881.	1914.	1847.	1899.
TOTAL MATERIAL RECOVERY, WT. %	99.7	99.6	100.2	100.3

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes
* - Catalyst included.

APPENDIX C

MATERIAL BALANCES FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5019	5020	5020A	5021
COAL TESTED	ILLINOIS NO. 6	PRINCE D	PRINCE D	LINGAN L
CATALYST TESTED	NM 502	NM 502	NM 502	NM 502
CATALYST, WT. % maf COAL	10	10	10	20
MOISTURE, WT. %	3.17	2.45	2.33	1.28
ASH, WT. % DRY BASIS	11.13	9.26	9.26	4.10
HOURS ON STREAM	6.05	6.08	6.00	8.00
DURATION OF BALANCE PERIOD, HRS.	1.99	2.02	2.01	2.02
INPUT (g):				
H_2O	17.7	13.5	12.7	7.2
Ash	60.2	49.7	49.2	22.6
maf Coal	481.	487.	482.	529.
Solvent	1288.	1266.	1253.	1230.
H_2	69.8	71.0	69.7	68.6
Catalyst	48.1	48.9	48.2	105.8
Total	1965.	1936.	1915.	1963.
OUTPUT (g):				
H_2O	66.9	68.5	47.6	37.2
*Ash	101.3	89.0	93.1	117.5
Organic THF Insolubles[1]	32.2	46.0	45.8	55.3
THF Solubles[2]	1659.	1625.	1651.	1632.
H_2	42.4	48.4	44.9	41.6
Other Gases	62.9	70.3	65.4	65.8
Total	1965.	1947.	1948.	1949.
TOTAL MATERIAL RECOVERY WT. %	100.0	100.6	101.7	99.3

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes
* - Catalyst included.

APPENDIX C

MATERIAL BALANCES FOR CATALYTIC HYDROGENATION EXPERIMENTS

RUN NO.	5022	5023	5024	5026
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NM 502	NM 502	NM 502	NM 502
CATALYST, WT. % maf COAL	20	20	20	10
MOISTURE, WT. %	1.28	1.28	1.28	1.28
ASH, WT. % DRY BASIS	4.10	4.10	4.10	4.10
HOURS ON STREAM	8.03	6.50	6.49	6.50
DURATION OF BALANCE PERIOD, HRS.	2.03	2.47	2.49	2.42
INPUT (g):				
H_2O	7.2	6.6	6.6	6.4
Ash	22.9	20.8	20.8	20.3
maf Coal	536.	486.	486.	475.
Solvent	1246.	1130.	1130.	1151.
H_2	68.9	65.3	65.9	63.9
Catalyst	107.3	97.2	97.2	47.4
Total	1988.	1806.	1807.	1764.
OUTPUT (g):				
H_2O	39.8	38.5	41.8	28.7
*Ash	116.5	107.1	102.7	59.8
Organic THF Insolubles[1]	55.1	44.3	45.2	39.4
THF Solubles[2]	1632.	1479.	1453.	1506.
H_2	42.0	30.1	33.6	33.5
Other Gases	82.3	86.2	87.0	87.8
Total	1968.	1785.	1763.	1755.
TOTAL MATERIAL RECOVERY, WT. %	99.0	98.8	97.6	99.5

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes
* - Catalyst included.

APPENDIX C

MATERIAL BALANCES FOR FIRST STAGE DISSOLUTION EXPERIMENTS

RUN NO.	5027	5028	5029	5030
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NONE	NONE	NONE	NONE
CATALYST, WT. % maf COAL	0.0	0.0	0.0	0.0
MOISTURE, WT. %	1.28	1.28	1.28	1.28
ASH, WT. % DRY BASIS	4.10	4.10	4.10	4.10
HOURS ON STREAM	6.09	6.03	6.05	6.03
DURATION OF BALANCE PERIOD, HRS	0.98	1.99	2.00	2.00
INPUT (g):				
H_2O	7.8	8.0	7.7	7.8
Ash	24.7	25.2	24.4	24.6
maf Coal	577.	589.	571.	575.
Solvent	1422.	1453.	1407.	1417.
H_2	43.7	45.2	37.8	45.4
Catalyst	0.0	0.0	0.0	0.0
Total	2075.	2120.	2048.	2070.
OUTPUT (g):				
H_2O	22.8	23.5	24.5	17.8
*Ash	24.0	23.1	22.5	24.4
Organic THF Insolubles[1]	95.9	87.5	92.5	102.0
THF Solubles[2]	1813.	1872.	1797.	1862.
H_2	37.7	40.2	35.4	43.3
Other Gases	71.5	100.0	102.3	42.9
TOTAL	2065.	2146.	2074.	2092.
TOTAL MATERIAL RECOVERY, WT. %	99.5	101.2	101.3	101.1

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes
* - Catalyst included.

APPENDIX C

MATERIAL BALANCES FOR FIRST STAGE DISSOLUTION EXPERIMENTS

RUN NO.	5031	5032	5033	5034
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED	NONE	NONE	NONE	NONE
CATALYST, WT. % maf COAL	0.0	0.0	0.0	0.0
MOISTURE, WT. %	1.28	1.28	1.28	1.28
ASH, WT. % DRY BASIS	4.10	4.10	4.10	4.10
HOURS ON STREAM	6.00	7.03	6.00	6.07
DURATION OF BALANCE PERIOD	2.00	0.99	1.00	2.02
INPUT (g):				
H_2O	8.2	7.6	7.9	8.0
Ash	26.1	24.1	25.1	25.4
maf Coal	610.	564.	586.	594.
Solvent	1503.	1390.	1445.	1463.
H_2	45.0	45.4	45.4	54.5
Catalyst	0.0	0.0	0.0	0.0
Total	2192.	2031.	2109.	2145.
OUTPUT (g):				
H_2O	23.6	20.5	19.7	21.9
*Ash	24.1	22.5	24.3	23.4
Organic THF Insolubles[1]	100.6	85.8	107.3	92.4
THF Solubles[2]	1935.	1789.	1869.	1913.
H_2	42.7	70.2	40.7	49.3
Other Gases	60.4	39.8	40.5	59.2
Total	2186.	2028.	2102.	2159.
TOTAL MATERIAL RECOVERY, WT. %	99.7	99.9	99.7	100.7

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes
* - Catalyst included.

APPENDIX C

MATERIAL BALANCES FOR NON-INTEGRATED TWO-STAGE LIQUEFACTION TESTS

RUN NO.	5035 I	5035 II	5036 I	5036 II
COAL TESTED	LINGAN L	LINGAN L	LINGAN L	LINGAN L
CATALYST TESTED		NM 502		NM 502
CATALYST, WT. % maf COAL		10.		20
MOISTURE, WT. %	1.28	1.28	1.28	1.28
ASH, WT. % DRY BASIS	4.10	4.10	4.10	4.10
HOURS ON STREAM	13.00	6.17	12.25	7.00
DURATION OF BALANCE PERIOD, HRS.	11.00	2.17	9.75	2.99
INPUT (g):				
H_2O	87.6	1.0	78.2	2.0
Ash	276.9	29.2	247.3	26.4
maf Coal	6,476.	683.	5,785.	617.
Solvent	15,960.	1,685.	14,258.	1,520.
H_2	499.	93.6	442.3	96.9
Catalyst	0.0	68.3	0.0	123.2
Total	23,300.	2,560.	20,811.	2,386.
OUTPUT (g):				
H_2O	243.9	6.2	217.3	23.7
*Ash	273.0	95.0	250.2	134.4
Organic THF Insolubles[1]	1,012.0	76.4	903.7	63.2
THF Solubles[2]	20,448.	2,280.	18,326.	2,028.
H_2	461.2	85.9	407.2	79.4
Other Gases	800.7	28.2	633.7	45.6
Total	23,239.	2,572.	20,758.	2,374.
TOTAL MATERIAL RECOVERY, WT. %	99.7	100.5	99.7	99.5

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes
* - Catalyst included.

APPENDIX C

MATERIAL BALANCES FOR NON-INTEGRATED TWO-STAGE LIQUEFACTION TESTS

RUN NO.	5037 I	5037 II
COAL TESTED	LINGAN L	LINGAN L
CATALYST TESTED		NM 502
CATALYST, WT. % maf COAL		20
MOISTURE, WT. %	1.28	1.28
ASH, WT. % DRY BASIS	4.10	4.10
HOURS ON STREAM	9.53	6.50
DURATION OF BALANCE PERIOD, HRS.	7.50	2.48
INPUT (g):		
H_2O	58.4	3.2
Ash	184.7	20.9
maf Coal	4,319.	490.
Solvent	10,645.	1,208.
H_2	340.2	75.1
Catalyst	0.0	98.1
Total	15,547.	1,895.
OUTPUT (g):		
H_2O	138.5	15.5
*Ash	180.3	106.4
Organic THF Insolubles[1]	699.1	51.9
THF Solubles[2]	14,011.	1,630
H_2	300.2	58.6
Other Gases	316.5	36.4
Total	15,646.	1,899.
TOTAL MATERIAL RECOVERY, WT. %	100.6	100.2

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes

* - Catalyst included.

APPENDIX D

ELEMENTAL ANALYSIS AND DISTILLATION DATA FOR PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM CONTINUOUS TESTS

ANALYSIS OF PENTANE AND TOLUENE SOLUBLE FRACTIONS FROM DONOR SOLVENT LIQUEFACTION TESTS

RUN NO.	5001	5002	5003	5004
Fraction	Toluene Soluble	Toluene Soluble	Toluene Soluble	Toluene Soluble
Carbon, Wt.%	89.88	90.43	90.51	90.69
Hydrogen, Wt.%	8.02	8.09	8.24	8.27
Nitrogen, Wt.%	0.35	0.36	0.37	0.38
Sulphur, Wt.%	0.218	0.224	0.071	0.070

Simulated Distillation Data, ^{o}C
(by Gas Chromatography)

Fraction	Pentane Soluble	Toluene Soluble	Pentane Soluble	Toluene Soluble	Pentane Soluble	Toluene Soluble	Pentane Soluble	Toluene Soluble
IBP/5	164/200	183/212	172/207	185/214	176/213	188/220	176/213	188/220
10/30	228/282	233/284	230/283	233/283	238/286	240/287	238/286	241/287
50	311	312	311	311	313	314	313	314
70/90	342/387	345/386	341/379	345/385	344/383	344/384	344/386	347/388
95/EP	418/474	414/483	407/465	414/483	413/476	416/480	416/478	419/481

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM CATALYTIC HYDROGENATION TESTS

RUN NO.	5011	5018	5020A	5023	5024
Pentane Soluble Fraction					
Carbon, Wt. %	90.86	90.43	90.54	89.79	90.02
Hydrogen, Wt. %	7.87	8.03	7.86	7.68	8.95
Nitrogen, Wt. %	0.48	0.44	0.41	0.44	0.26
Sulphur, Wt. %	0.051	0.067	0.063	0.023	0.014
Oxygen, Wt. %					0.60
Toluene Soluble Fraction					
Carbon, Wt. %	90.20	89.77	90.42	88.23	90.10
Hydrogen, Wt. %	7.61	7.88	7.61	7.49	8.47
Nitrogen, Wt. %	0.60	0.50	0.52	0.53	0.35
Sulphur, Wt. %	0.060	0.099	0.084	0.030	0.021
Oxygen, Wt. %		1.29	1.30		1.01
THF Soluble Fraction					
Carbon, Wt. %	90.31	89.68	89.33	88.76	89.18
Hydrogen, Wt. %	7.64	7.90	7.64	7.54	8.50
Nitrogen, Wt. %	0.66	0.51	0.53	0.58	0.38
Sulphur, Wt. %	0.071	0.115	0.085	0.030	0.019
Oxygen, Wt. %					1.35

Lingan coal (Batch L) used for Run Nos. 5011, 5023 and 5024.
Illinois No. 6 coal used for Run No. 5018.
Prince coal (Batch D) used for Run No. 5020A.

DISTILLATION DATA ON PENTANE AND TOLUENE SOLUBLE FRACTIONS
FROM CATALYTIC HYDROGENATION TESTS

RUN NO.	5011	5018	5020A	5023	5024
Distillation Data, °C (by Gas Chromatography)					
Pentane Soluble Fraction					
IBP/5	180/211	156/209	103/208	114/208	104/198
10/30	237/287	233/286	234/286	219/278	213/268
50	317	315	315	313	302
70/90	344/387	344/387	344/386	340/379	328/377
95/EP	418/482	412/475	413/476	411/478	409/478
Toluene Soluble Fraction					
IBP/5	203/220	192/219	196/225	190/213	190/212
10/30	242/287	243/287	248/288	237/286	234/275
50	316	316	316	314	305
70/90	347/390	346/388	347/387	342/382	330/377
95/EP	418/482	414/477	414/478	412/477	413/481

Lingan coal (Batch L) used for Run Nos. 5011, 5023 and 5024.
Illinois No. 6 coal used in Run No. 5018.
Prince coal (Batch D) used in Run No. 5020A.

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS
FROM NON-INTEGRATED TWO-STAGE LIQUEFACTION TESTS

RUN NO.	5028	5036 I	5036 II	5037 I	5037 II
Pentane Soluble Fraction					
Carbon, Wt. %	89.26	90.35	90.18	89.34	89.32
Hydrogen, Wt. %	7.88	8.00	8.32	8.45	8.65
Nitrogen, Wt. %	0.24	0.22	0.29	0.19	0.28
Sulphur, Wt. %	0.025	0.026	0.011	0.027	0.016
Toluene Soluble Fraction					
Carbon, Wt. %	90.08	89.65	88.61	89.22	89.47
Hydrogen, Wt. %	7.57	7.90	8.17	8.30	8.27
Nitrogen, Wt. %	0.41	0.44	0.41	0.34	0.42
Sulphur, Wt. %	0.049	0.056	0.025	0.059	0.027
THF Soluble Fraction					
Carbon, Wt. %	89.22	89.54	89.15	89.33	89.32
Hydrogen, Wt. %	7.45	7.87	8.14	8.05	8.26
Nitrogen, Wt. %	0.57	0.55	0.45	0.54	0.48
Sulphur, Wt. %	0.053	0.076	0.029	0.092	0.032

Lingan coal (Batch L) used in above Runs.

DISTILLATION DATA ON PENTANE AND TOLUENE SOLUBLE FRACTIONS
FROM NON-INTEGRATED TWO-STAGE LIQUEFACTION TESTS

RUN NO.	5028	5036 I	5036 II	5037 I	5037 II
Distillation Data, °C (by Gas Chromatography)					
Pentane Soluble Fraction					
IBP/5	102/208	181/214	173/212	186/224	183/219
10/30	233/282	239/287	237/283	247/289	241/286
50	313	314	311	314	311
70/90	340/379	342/381	340/385	341/381	338/379
95/EP	413/478	414/478	419/482	412/477	413/481
Toluene Soluble Fraction					
IBP/5	192/221	206/232	203/227	205/233	203/229
10/30	241/287	249/288	245/286	249/289	248/287
50	316	316	312	314	313
70/90	348/386	349/388	340/383	343/387	340/386
95/EP	418/479	418/478	417/481	416/479	418/482

Lingan coal (Batch L) used in above Runs.

APPENDIX E

ANALYSIS OF DISTILLATE PRODUCT FROM SEPARATOR BOTTOMS

ANALYSIS OF DISTILLATE PRODUCT FROM SEPARATOR BOTTOMS

RUN NO.	5011	5018	5019	5020A	5023	5024	5037II
Specific Gravity, 60/60°F	1.051	1.048	1.053	1.056	1.046	1.016	1.032
Viscosity, cSt @ 40°C	7.75	8.56	8.34	8.75	7.22	5.60	7.82
Conradson Carbon, Wt.%	0.19	0.28	0.28	0.28	0.21	0.17	0.15
Carbon, Wt.%	89.43	89.68	89.30	90.13	89.12	89.60	89.66
Hydrogen, Wt.%	7.66	8.04	7.97	7.91	7.80	9.00	8.85
Nitrogen, Wt.%	0.46	0.42	0.41	0.41	0.43	0.23	0.26
Sulphur, Wt.%	0.042	0.032	0.031	0.052	0.019	0.003	0.008
Oxygen, Wt.%		1.39		1.08		0.95	
Proton Distribution %							
H_{ar} (9-6 ppm)	38.95	31.20	33.05	34.66	39.65	22.98	25.73
H_{α} (3.5-2 ppm)	30.45	33.13	32.04	31.83	28.55	31.00	30.26
H_{β} (2-1.1 ppm)	25.14	26.88	28.40	29.61	25.71	38.55	34.56
H_{γ} (1.1-0.4 ppm)	5.52	8.79	6.51	3.90	6.09	7.48	9.45
Distillation Data, °C (by Gas Chromatography)							
IBP/5	164/209	167/209	172/209	181/212	163/208	159/208	184/217
10/30	227/279	233/286	234/286	238/287	226/278	220/269	241/286
50	313	314	314	316	312	299	312
70/90	338/379	341/380	341/380	342/382	338/378	323/372	337/378
95/EP	409/479	408/476	408/475	412/478	408/479	404/478	407/476

APPENDIX F

RESULTS OF HPLC APPLIED TO COAL-DERIVED LIQUIDS

APPENDIX F

RESULTS OF HPLC APPLIED TO COAL-DERIVED LIQUIDS

Chromatographic methods are widely used for the separation and characterization of products obtained from crude oil and coal. Coal liquids, however, contain a larger variety of compounds than petroleum oils. Some separation schemes originally developed for petroleum heavy ends have been applied to coal liquids. During Phase V an open column method (ASTM D-2007) and a high performance liquid chromatography (HPLC) method were used for the separation of coal liquids into saturates, aromatics and polar compounds.

Lumpkin and Aczel (1) used a modified version of the clay-gel adsorption chromatographic method, ASTM D-2007, to separate fractions boiling above the naphtha range into saturate, aromatic and polar fractions. More recently, several workers (2-5) reported the use of HPLC for the separation of polycyclic aromatic hydrocarbons (PAH).

In the HPLC method presented here a Perkin-Elmer analytical amino-cyano column was used to separate coal liquids into saturates, aromatics and polar compounds. Furthermore, separation into *double bond* fractions was achieved. A number of model compounds were used to define the specific regions of the chromatograph as shown in Figure 11. The regions were defined by the number of

double bonds in a given hydrocarbon molecule ranging from 0 (saturates) to 8 (tetracyclic PAH).

The separation was achieved on a 10 µL sample. Pentane at a flowrate of 1 mL/minute was used as the mobile phase in a forward operation then the column was backflushed with methyl-t-ether at a flowrate of 4 mL/minute. Forward eluting compounds were monitored by differential refractometry, while backflushed components were detected by the UV detector.

After establishing separation cut-points between 0, 3, 5, 6, 7 and 8 *double bond* fractions the separation procedure was applied to coal liquids. Individual fractions were collected at the exit part of the detector. Three injections of each sample were made and the respective fractions from each run were combined. The fractions were concentrated carefully in preweighed vials, dried with nitrogen and weighed. Elution profiles for hydrogenated anthracene oil and distillate samples from continuous coal liquefaction experiments are shown in Figures 12 - 17.

Results obtained using both separation procedures are presented in Table 15. Excellent recovery was achieved in both methods. Agreement between the methods ranged from fair to good. A comparison of HAO 55 and HAO 56 illustrates that the 9% H oil has a higher level of saturates and lower weight percentages of 6, 7 and 8 *double bond* fractions than the 7.5% H oil. The results also show that the dis-

tillate from each run is higher in saturates and polar compounds than the vehicle oil used.

Although the analytical potential of this HPLC separation technique based on the use of an amino-cyano column has not been confirmed by detailed analysis of the fractions, other workers have done so for similar HPLC methods. In future work HPLC will be used to monitor hydrocarbon groups in start-up oil, recycle oil and distillate products.

REFERENCES

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Figure 11 - HPLC ELUTION PROFILES FOR MODEL COMPOUNDS

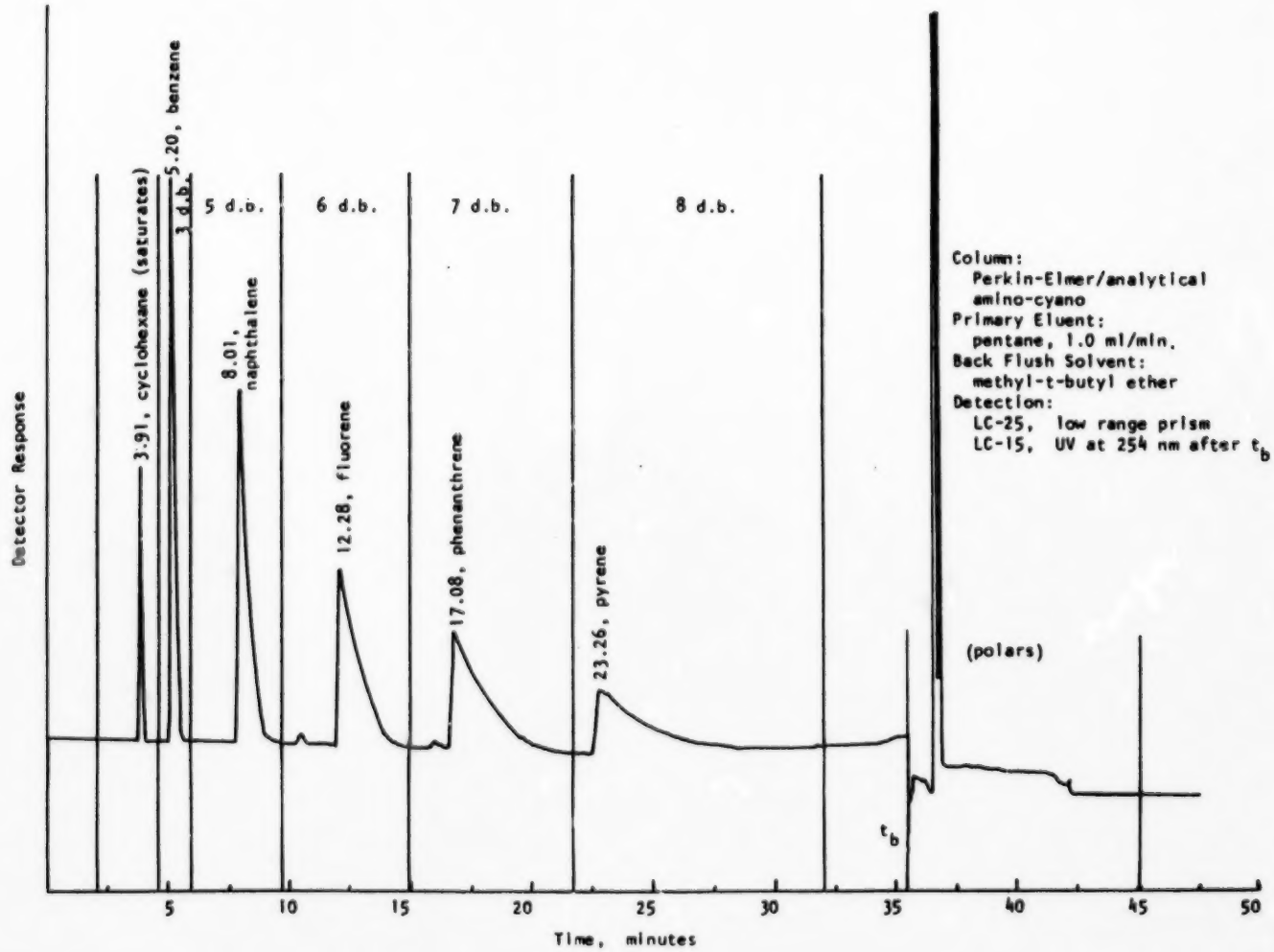


Figure 12 - HPLC SEPARATION OF HYDROGENED ANTHRACENE OIL WITH 7.54% WT. % HYDROGEN

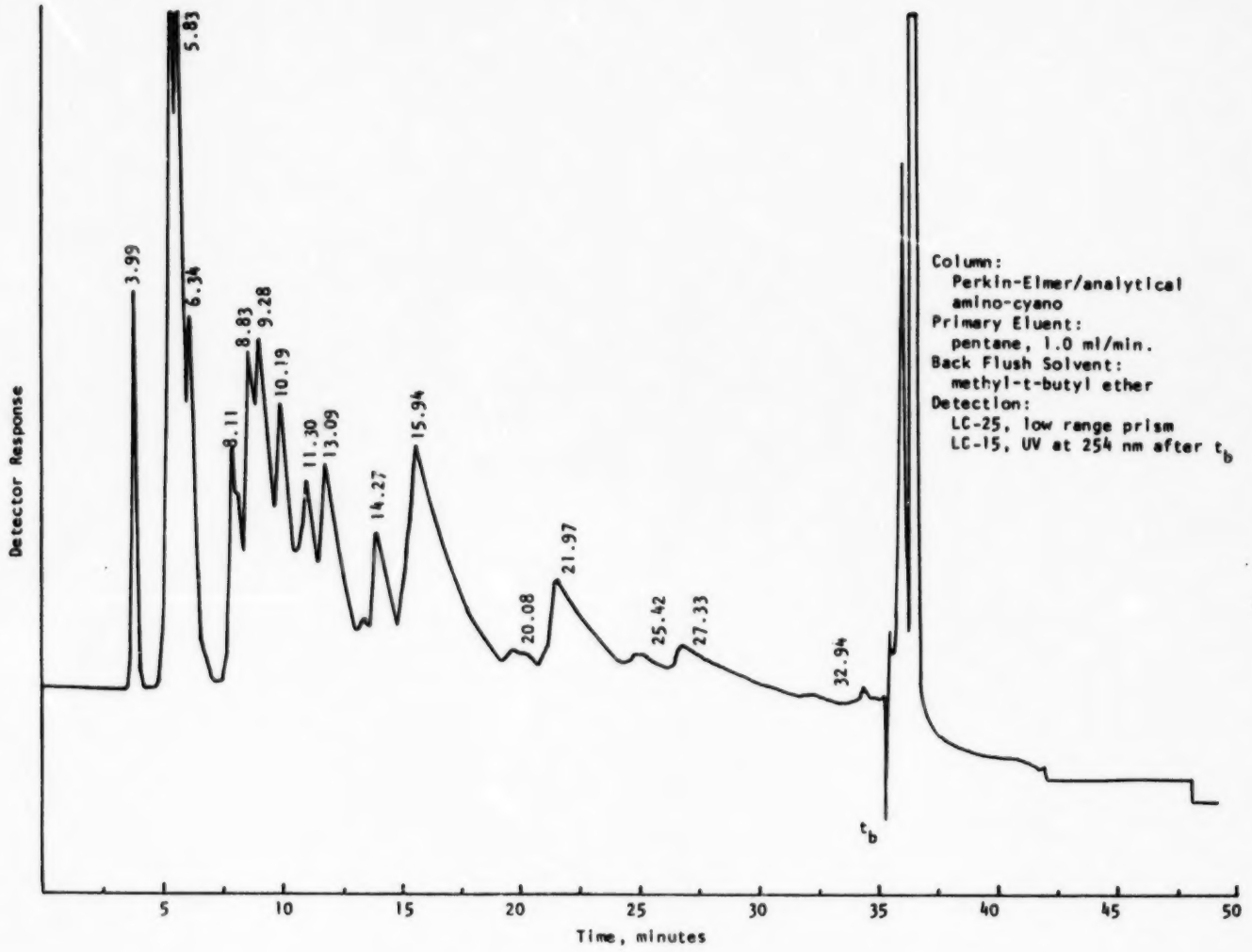


Figure 13 - HPLC SEPARATION OF HYDROGENATED ANTHRACENE OIL WITH 9.01 WT. % HYDROGEN

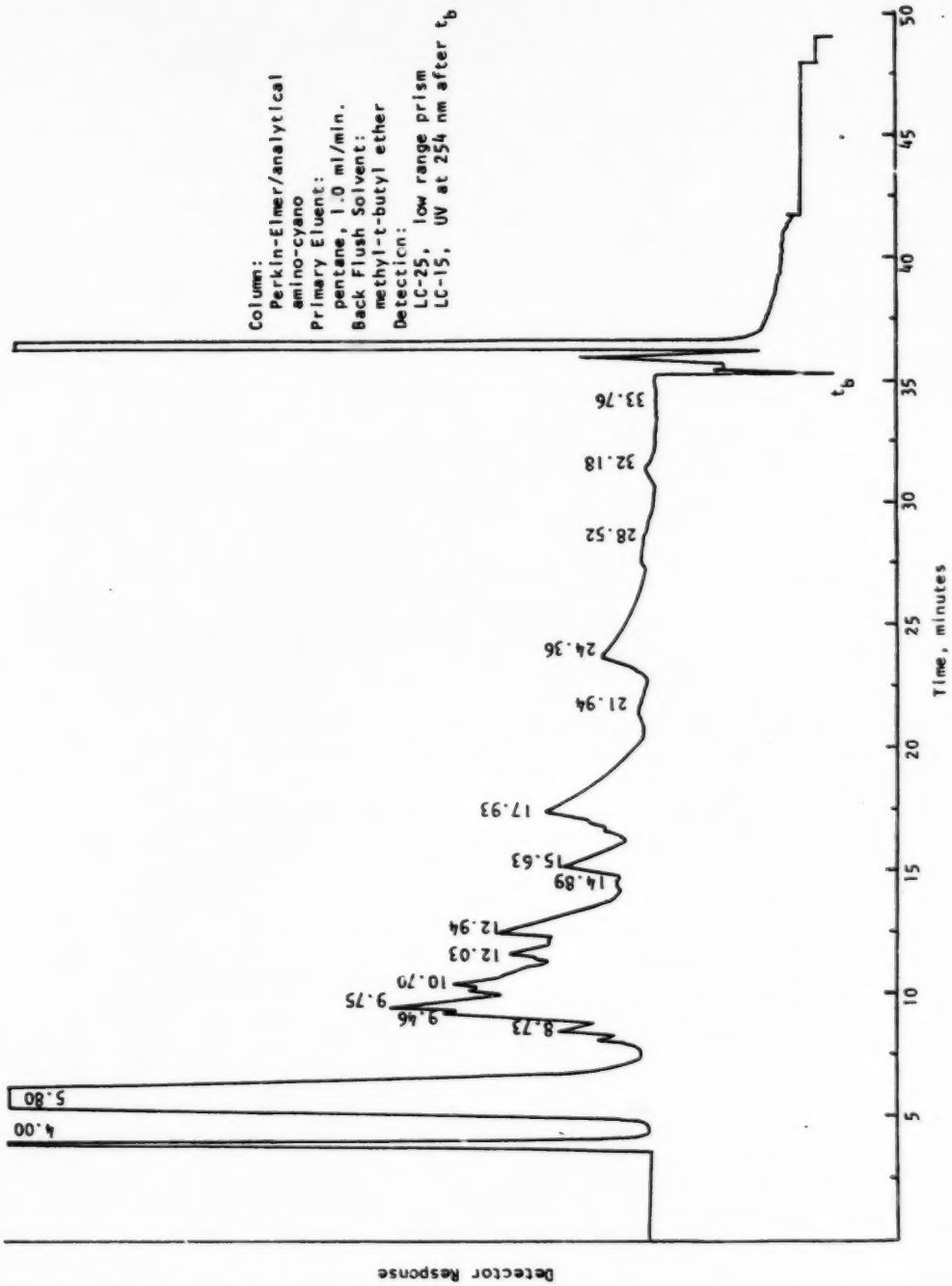


Figure 14 - HPLC SEPARATION OF DISTILLATE FROM RUN NO. 5011

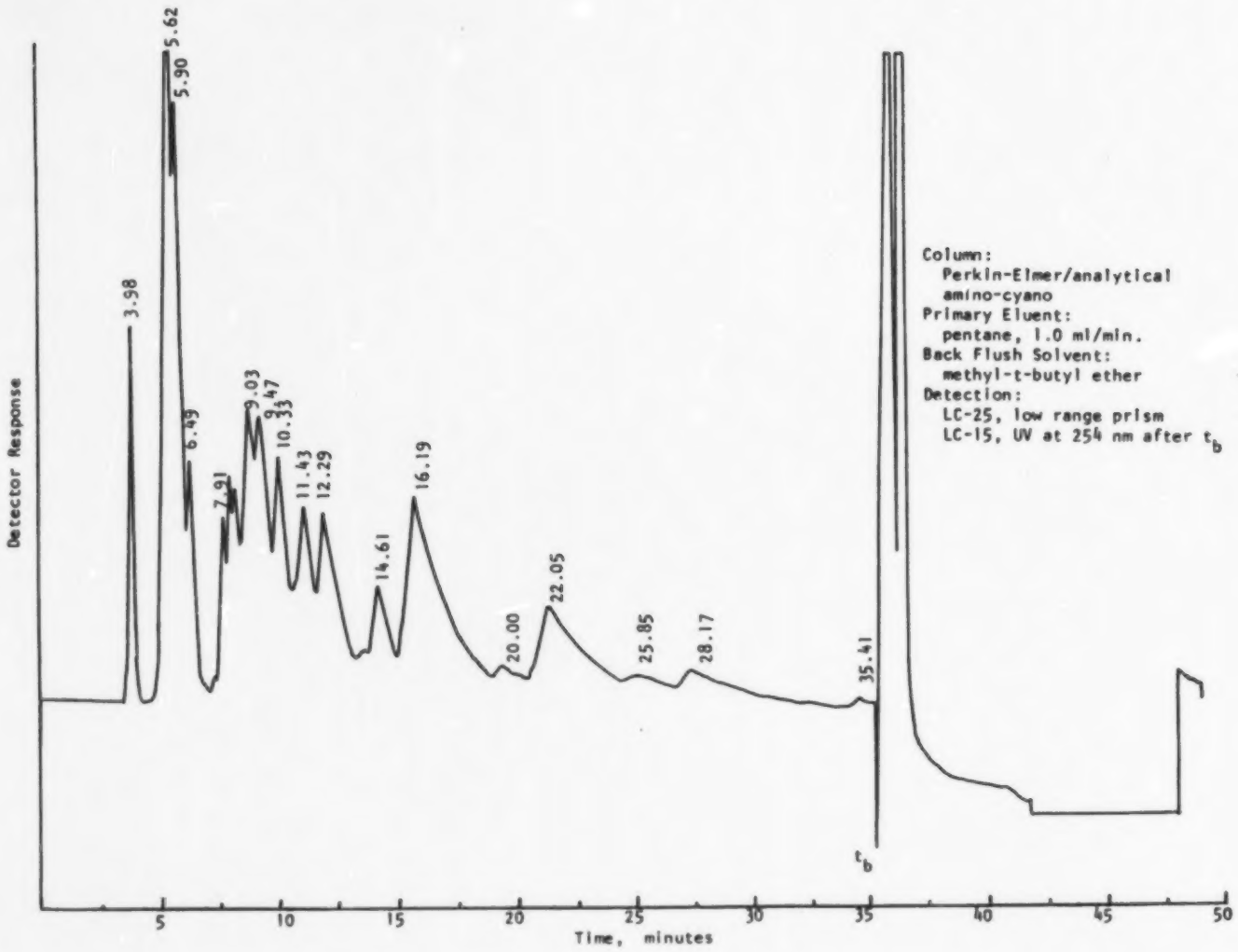
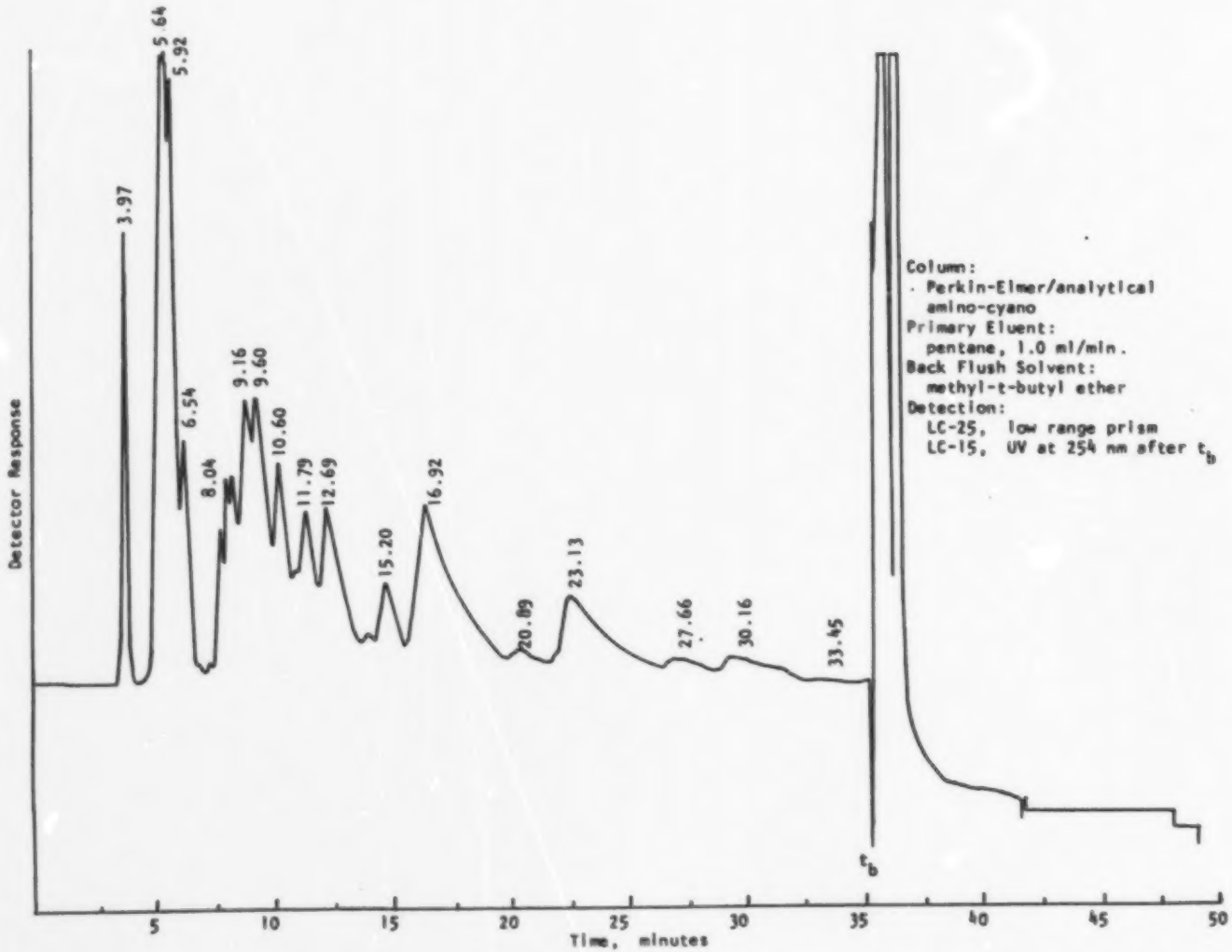


Figure 15 - HPLC SEPARATION OF DISTILLATE FROM RUN NO. 5018



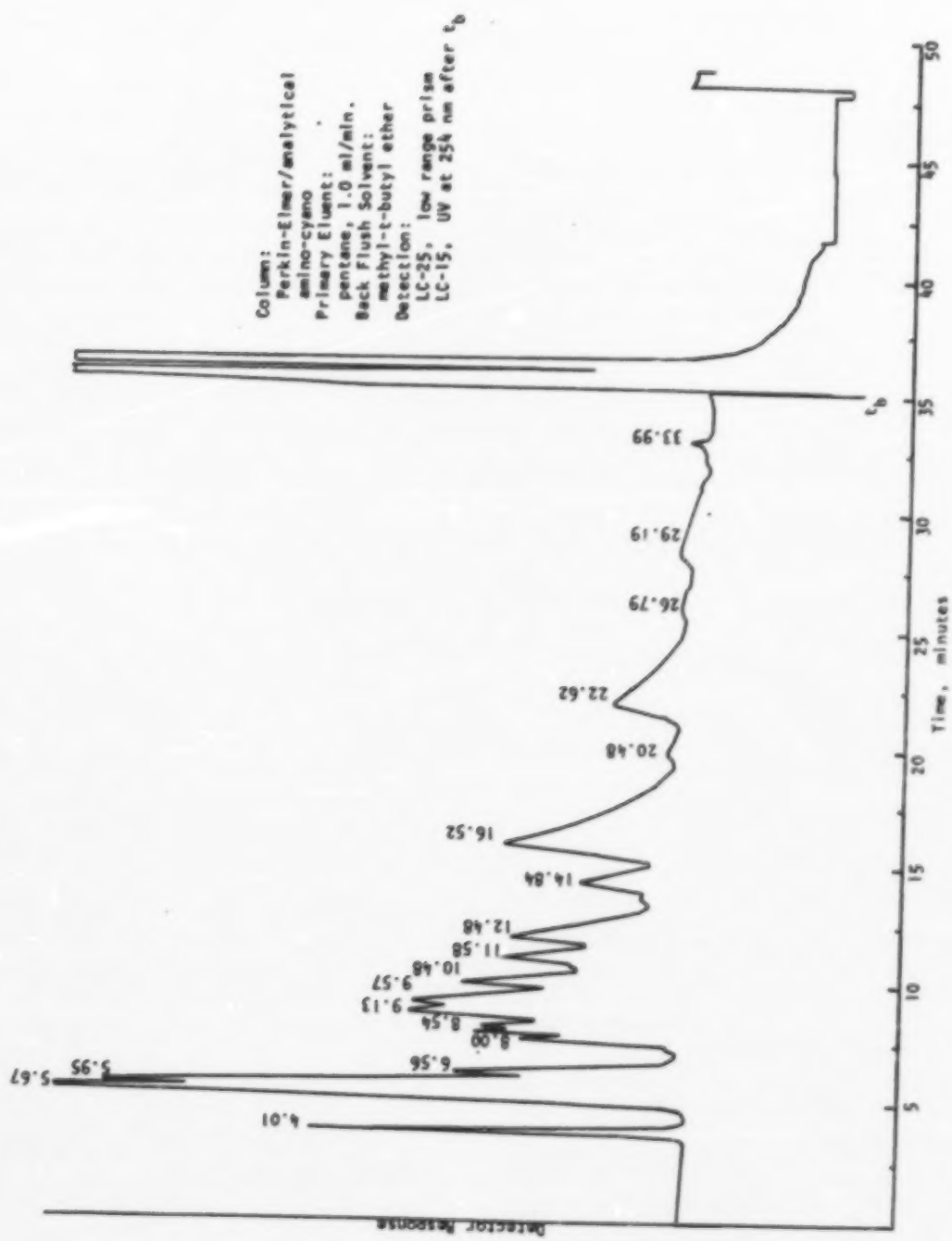


Figure 16 - HPLC SEPARATION OF DISTILLATE FROM RUN NO. 5020A

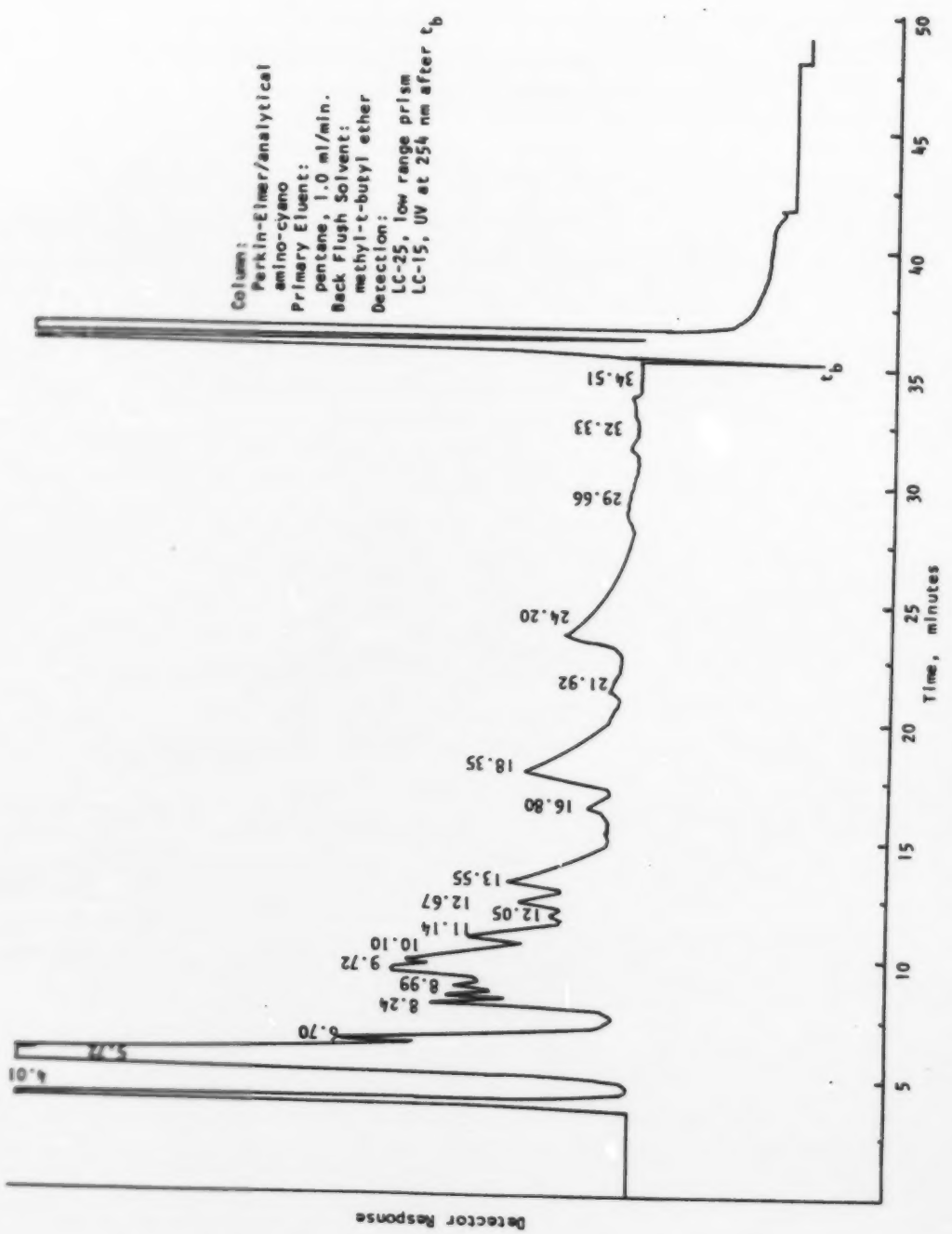


Figure 17 - HPLC SEPARATION OF DISTILLATE FROM RUN NO. 5024

TABLE 15

CHROMATOGRAPHIC SEPARATION OF HYDROGENATED ANTHRACENE OIL AND DISTILLATE FRACTIONS FROM SELECTED CONTINUOUS COAL LIQUEFACTION EXPERIMENTS

SAMPLE DESCRIPTION	HAO 55		HAO 56		RUN NO. 5011 DISTILLATE		RUN NO. 5024 DISTILLATE		RUN NO. 5020A DISTILLATE		RUN NO. 5018 DISTILLATE	
SEPARATION METHOD	HPLC	ASTM D-2007	HPLC	ASTM D-2007	HPLC	ASTM D-2007	HPLC	ASTM D-2007	HPLC	ASTM D-2007	HPLC	ASTM D-2007
Saturates, Wt. %	2.60	3.42	12.49	11.72	3.62	3.68	14.58	12.35	4.41	3.81	4.71	5.04
Aromatics, Wt. %	82.49	86.06	81.40	85.62	82.68	84.18	78.45	80.32	80.39	82.92	81.82	83.35
3 double bonds	7.01		11.63		9.15		13.80		7.84		8.12	
5 double bonds	20.70		27.91		24.51		23.91		22.22		21.75	
6 double bonds	25.48		20.60		19.93		19.19		18.63		20.78	
7 double bonds	16.56		9.63		14.38		11.45		16.34		16.88	
8 double bonds	12.74		11.63		14.71		10.10		15.36		14.29	
Polars, Wt. %	12.42	10.53	6.31	3.00	14.38	12.06	9.43	7.75	15.69	13.48	13.96	11.88
Recovery, Wt. %	97.51	100.01	100.20	100.34	100.68	99.92	102.46	100.42	100.49	100.21	100.49	100.27

Lingan harbor coal used in Run Nos. 5011, 5024.
Prince hub coal used in Run No. 5020A.
Illinois No. 6 coal used in Run No. 5018.

APPENDIX G

CALCULATIONS FOR SIMPLEX SELF-DIRECTING EVOLUTIONARY OPERATION

SIMPLEX SELF-DIRECTING EVOLUTIONARY OPERATION (SSEVOP)

The process variables evaluated were as follows:

x_1 Hydrogen Treatrate, Wt. % dry coal
x_2 Catalyst Concentration, Wt. % maf coal
x_3 Temperature, ^{o}C
x_4 Slurry Feedrate, kg/hr

The mathematical expression is of the form:

$$y = a_0 + a_1x_1 + a_2x_2 + \ldots + a_nx_n$$

The average level and range of each factor is shown below:

	x_1	x_2	x_3	x_4
Mean Level	13	8	455	1.000
Standard Deviation	2	1.5	8	0.200
Total Shift	4	3	16	0.400

The simplex design matrix is of the form:

Run Number	x_1	x_2	x_3	x_4
1	$-a_1$	$-a_2$	$-a_3$	$-a_4$
2	a_1	$-a_2$	$-a_3$	$-a_4$
3	0	$2a_2$	$-a_3$	$-a_4$
4	0	0	$3a_3$	$-a_4$
5	0	0	0	$4a_4$

Using the total shift values, let:

$2a_1 = 4$ $\therefore$ $a_1 = 2$

$3a_2 = 3$ $a_2 = 1$ $2a_2 = 2$

$4a_3 = 16$ $a_3 = 4$ $3a_3 = 12$

$5a_4 = 0.400$ $a_4 = 0.080$ $4a_4 = 0.320$

Design matrix in actual units:

Run No.	H_2 Treatrate	Catalyst Conc.	Temperature	Slurry Feedrate
1	11	7	451	0.920
2	15	7	451	0.920
3	13	10	451	0.920
4	13	8	467	0.920
5	13	8	455	1.320

SSEVOP CALCULATIONS

RUN NO.	HYDROGEN TREATRATE WT. %	CATALYST LEVEL WT. %	TEMPERATURE °C	SLURRY FEEDRATE kg/hr	RESPONSES	
					OIL WT. %	UNREACTED COAL, WT. %
5009	11	7	451	0.920	36.5	8.6
5010	15	7	451	0.920	34.7	8.4
5011	13	10	451	0.920	36.1	8.8
5012	13	8	467	0.920	34.6	7.9
5014	~~13~~	~~8~~	~~455~~	~~1.320~~	33.1	9.5
Sum	52	32	1820	3.600		
Average	13	8	455	0.920		
2X Average (-)	26	16	910	1.840		
Discard Point	13	8	455	1.320		
New Point	13	8	455	0.520		

APPENDIX H

STATISTICAL ANALYSIS OF 2^3 FACTORIAL EXPERIMENT ON LINGAN HARBOUR SEAM COAL

APPENDIX H

RESPONSE FACTOR: GAS MAKE

SLURRY FEEDRATE	440°C		460°C	
	6.89 MPa	10.34 MPa	6.89 MPa	10.34 MPa
1 kg/hr	12.5	12.3	20.9	19.6
2 kg/hr	8.9	9.2	15.0	14.7

$\Sigma x = 113.1 \qquad C = \frac{(\Sigma x)^2}{n} = \frac{(113.1)^2}{8} = 1{,}598.9513$

$SS_{Total} = \Sigma x^2 - C = 134.4987$

$SS_{Temperature} = 6{,}768.45/4 - C = 93.1612$

$SS_{Pressure} = 6{,}396.93/4 - C = 0.2812$

$SS_{Feedrate} = 6{,}548.93/4 - C = 38.2812$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	93.1612	93.1612	134.2767	.99	+ 27.3
Pressure	1	0.2812	0.2812	0.4053	.75	- 1.5
Feedrate	1	38.2812	38.2812	55.1761	.99	- 17.5
Residual	4	2.7751	0.6938			
Total	7	134.4987				

* n = 8

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 113.1$

$\bar{x} = \frac{\Sigma x}{n} = \frac{113.1}{8} = 14.1$

$S^2 = \frac{SS}{df} = \frac{2.7751}{4} = 0.6938$

$S = \sqrt{0.6938} = 0.83$

$S_{\bar{x}} = \frac{S}{\sqrt{n}} = \frac{0.83}{\sqrt{8}} = 0.3$

∴ the mean of 8 results at the 2σ confidence interval is:

14.1 ± 0.6

APPENDIX H

RESPONSE FACTOR: OIL

SLURRY FEEDRATE	440°C		460°C	
	6.89 MPa	10.34 MPa	6.89 MPa	10.34 MPa
1 kg/hr	19.5	19.7	17.8	18.9
2 kg/hr	16.4	14.5	20.0	21.3

$\Sigma x = 148.1$ $\quad C = \frac{(\Sigma x)^2}{n} = \frac{(148.1)^2}{8} = 2{,}741.7013$

$SS_{Total} = \Sigma x^2 - C = 33.5887$

$SS_{Temperature} = 10{,}998.01/4 - C = 7.8012$

$SS_{Pressure} = 10{,}967.05/4 - C = 0.0612$

$SS_{Feedrate} = 10{,}973.65/4 - C = 1.7112$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	7.8012	7.8012	1.2994	.75	+ 7.9
Pressure	1	0.0612	0.0612	0.0102	.75	+ 0.7
Feedrate	1	1.7112	1.7112	0.2850	.75	- 3.7
Residual	4	24.0151	6.0038			
Total	7	33.5887				

* n = 8

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 148.1$

$\bar{x} = \frac{\Sigma x}{n} = \frac{148.1}{8} = 18.5$

$S^2 = \frac{SS}{df} = \frac{24.0151}{4} = 6.0038$

$S = \sqrt{6.0038} = 2.45$

$S_{\bar{x}} = \frac{S}{\sqrt{n}} = \frac{2.45}{\sqrt{8}} = 0.9$

∴ the mean of 8 results at the 2σ confidence interval is:

18.5 ± 1.8

APPENDIX H

RESPONSE FACTOR: ASPHALTENES

SLURPY FEEDRATE	440°C		460°C	
	6.89 MPa	10.34 MPa	6.89 MPa	10.34 MPa
1 kg/hr	27.3	28.3	26.8	26.7
2 kg/hr	25.2	27.1	25.8	25.7

$\Sigma x = 212.9 \qquad C = \frac{(\Sigma x)^2}{n} = \frac{(212.9)^2}{8} = 5{,}665.8013$

$SS_{Total} = \Sigma x^2 - C = 7.0887$

$SS_{Temperature} = 22{,}667.41/4 - C = 1.0512$

$SS_{Pressure} = 22{,}666.85/4 - C = 0.9112$

$SS_{Feedrate} = 22{,}677.25/4 - C = 3.5112$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	1.0512	1.0512	2.6033	.75	- 2.9
Pressure	1	0.9112	0.9112	2.2566	.75	+ 2.7
Feedrate	1	3.5112	3.5112	8.6954	.95	- 5.3
Residual	4	1.6151	0.4038			
Total	7	7.0887				

* n = 8

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 212.9$

$\bar{x} = \frac{\Sigma x}{n} = \frac{212.9}{8} = 26.6$

$S^2 = \frac{SS}{df} = \frac{1.6151}{4} = 0.4038$

$S = \sqrt{0.4038} = 0.64$

$S_{\bar{x}} = \frac{S}{\sqrt{n}} = \frac{0.64}{\sqrt{8}} = 0.2$

∴ the mean of 8 results at the 2σ confidence interval is:

26.6 ± 0.4

APPENDIX H

RESPONSE FACTOR: PREASPHALTENES

SLURRY FEEDRATE	440°C		460°C	
	6.89 MPa	10.34 MPa	6.89 MPa	10.34 MPa
1 kg/hr	24.1	24.1	18.3	19.4
2 kg/hr	31.2	31.5	22.6	23.1

$\Sigma x = 194.3$ $\quad C = \frac{(\Sigma x)^2}{n} = \frac{(194.3)^2}{8} = 4{,}719.0613$

$SS_{Total} = \Sigma x^2 - C = 163.8700$

$SS_{Temperature} = 19{,}254.37/4 - C = 94.5312$

$SS_{Pressure} = 18{,}878.05/4 - C = 0.4512$

$SS_{Feedrate} = 19{,}129.37/4 - C = 63.2812$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	94.5312	94.5312	67.4452	.99	- 27.5
Pressure	1	0.4512	0.4512	0.3219	.75	+ 1.9
Feedrate	1	63.2812	63.2812	45.1493	.99	+ 22.5
Residual	4	5.6064	1.4016			
Total	7	163.8700				

* n = 8

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 194.3$

$\bar{x} = \frac{\Sigma x}{n} = \frac{194.3}{8} = 24.3$

$S^2 = \frac{SS}{df} = \frac{5.6064}{4} = 1.4016$

$S = \sqrt{1.4016} = 1.18$

$S_{\bar{x}} = \frac{S}{\sqrt{n}} = \frac{1.18}{\sqrt{8}} = 0.4$

∴ the mean of 8 results at the 2σ confidence interval is:

24.3 ± 0.8

APPENDIX I

OPERATING PROCEDURE FOR CONTINUOUS COAL LIQUEFACTION UNIT

PREPARATION OF UNIT PRIOR TO ACTUAL OPERATION

This procedure is normally carried out the day before a run.

1. Prepare V-13 product withdrawal line for venting. All valves except 1, 103, 118 and 122 are closed.

2. Open valves 7, 20, 21, 24 and 25 to depressurize the system.

3. Prepare V-13 product withdrawal line for vacuum.

4. Connect 2 L pressure vessel to valve 6. Attach vacuum gauge to pressure vessel and open adjacent valve, 6A. Open valve 6 and turn on the vacuum pump.

5. After one hour, close valves 24 and 25. Observe gauge reading for 5 minutes. Close valve 6 and 6A and then remove vacuum gauge. Shut off the vacuum pump.

6. Open valve 2 and add hydrogen to 13.78 MPa, close valve 2. Leave for a few hours and observe for pressure leaks.

7. Set the forward pressure regulator and the back pressure regulator (B.P.R.) to the desired settings.

8. Add high pressure hydrogen to the system through valve 2 to 4.83 MPa (700 psig) less than the desired pressure, close valve No. 2.

9. Turn on the cooling water to the magnedrive stirrers and check water hose for tightness of fit.

10. Set the furnace controller at 1100°F and the zone controllers at 55%. Set the reactor controller at 11°F above target setpoint.

11. Set the automatic timer to activate the STR furnace temperature controller at 5 hours prior to anticipated start-up (normally 5 a.m.). Turn the STR temperature controller switch to the 'ON' position and ensure it is not activated.

12. Set the temperature controller for high pressure separator No. 1, V-3, at 300°C.

13. Set the automatic timer to activate V-3 temperature controller at 4 hours prior to anticipated start-up. Turn the controller switch to the 'ON' position and ensure it is not activated.

ACTUAL OPERATION

Start-Up

1. Gradually increase the furnace controller to the desired level. DO NOT allow the temperatures of the STR to exceed 250°F per hour.

2. Turn on heating tapes on slurry transfer lines.

   No. 1 → Full (9 o'clock)

   No. 2 → 33% (3 o'clock) then increase to 66% (6 o'clock) one hour prior to slurry entering transfer lines

   No. 3 → 33% (3 o'clock) then increase to 66% (6 o'clock) one hour prior to slurry entering transfer lines

3. Turn on heating tape on high pressure separator No. 2, V-4.

   Separator 2 → Setting of 180

4. Turn on the condenser cooling water.

5. Activate K-Ray electronics for liquid level measurement system.

6. After one hour, open window on radioactive source.

7. Turn on the Hydrogen Flow Control System twenty minutes prior to run.

   Procedure

   a) Turn on the power.
   b) Open valve 3.
   c) Set the air pressure at 138 kPa (20 psig).
   d) Adjust the setpoint knob until the ball is in the center, then switch from manual to automatic.
   e) Set the pen to the desired value.
   f) Open valve 104 and close valve 103.
   g) Allow the Hydrogen Flow Control System to stabilize.
   h) Monitor exit gas flow rate for 15 minutes.

8. Open valve 101 and purge feed tank with nitrogen 15 minutes prior to slurry addition.

9. Charge V-1 with the slurry. Open valve 102 and turn on the circulating pump.

10. Turn off the circulating pump for 15 seconds and then turn circulating pump back on. This fills void spaces or gas pockets in the pump with slurry and ensures proper weight readings on the toledo scale.

11. When the temperature of the STR is at the desired level, purge the high pressure metering pump with slurry until the liquid is gas free.

Run

12. Open the plant valve (valve 5) and activate the high pressure metering pump.

13. Turn on the V-2 and V-3 magnedrive stirrers.

14. Monitor the pressures, temperatures, slurry feedrate and vent gas flow rates throughout the run.

15. A rapid increase in the reactor temperature will occur 15 to 20 minutes after the high pressure metering pump has been activated. When the reactor temperature reaches 25°F below the target temperature turn the furnace temperature controller down until the red light goes off. Increase the furnace temperature controller back to the original setpoint when the STR temperature levels off.

16. Open valves 106 and 108 10 minutes prior to Step 17 and purge V-7 with nitrogen.

17. After 1.4 kg of slurry has been pumped, open valves 111, 110, and 14.

18. Set K-5 for the automatic letdown (ALD) system to maintain a constant level in V-3. Connect plugs to ATO valves 11 and 12.

19. Open valve 9.

20. Set the selector switch on "Automatic During High Alert" and activate the automatic letdown system.

21. After the first ALD cycle, close valves 106 and 108.

22. Sample vent gas streams one-half hour prior to steady state sampling period.

23. Drain liquid from the separator overheads knock-out (V-13) periodically and 3 minutes prior to steady state sampling period by opening and closing valves 24 and 25 sequentially.

24. To initiate the steady state sampling period, record the toledo scale weight after valve "A" has de-energized on the final ALD cycle of the slop period.

25. Record the wet test meter reading and the circulating pump pan weight.

26. Open valves 109 and 13. Close valves 14 and 111.

27. Open valve 117 to drain V-12. Close valve 117.

28. Open valves 106 and 107 and purge V-6 with nitrogen.

29. On the next ALD cycle, the first in the steady state sampling period, mark the stroke made on the Hydrogen Flow Recorder.

30. Close valves 106 and 107.

31. Sample vent gas streams 15 minutes prior to end of steady state sampling period.

32. Three minutes prior to end of run, drain liquid from separator overheads knock-out (V-13) by opening and closing valves 24 and 25 sequentially.

33. At the end of the period, record the toledo scale weight after valve "A" has de-energized on the final ALD cycle of the sampling period. Close valve 9 and deactivate the automatic letdown system.

34. Record the wet test meter reading and the circulating pump pan weight.

35. Turn off the STR furnace temperature controller and the high pressure separator heaters. Reduce voltage to heating tapes on slurry transfer lines.

36. Shut off the Hydrogen Flow Control System.

Procedure

a) Turn the regulator knob until the ball is in the centre.
b) Switch from automatic to manual.
c) Shut off the air flow.
d) Close valve 3.
e) Turn off the power.

37. De-activate the high pressure metering pump.

38. Close the plant valve (valve 5) and valves 7 and 20.

39. Transfer contents of V-2 into an evacuated pressure vessel through valve 6. Leave valve 6 open for 1 hour.

40. Repeat Step 27.

41. Open valve 112. Manually withdraw slurry product from V-3 by opening valves 15 and 10, close valves 10 and 15. After 20 minutes and 60 minutes repeat this procedure. Close valve 112.

42. After one hour, repeat Step 27.

43. De-activate K-Ray electronics for liquid level measurement system.

44. Close window on radioactive source.

45. De-activate the circulating pump and drain slurry from the feed tank.

46. Open valves 20 and 21 to equalize the pressure.

47. Close valve 6 and open valve 2 to add high pressure hydrogen to the STR until its pressure is 10.34 MPa. After 15 minutes transfer contents of V-2 into evacuated pressure vessel through valve 6. Close valve 6.

48. Open valve 2 and add high pressure hydrogen to the STR until its pressure is 689 kPa (100 psig) in excess of the downstream pressure and then close valve 2.

49. Open valve 7 and equalize the pressure throughout the system. Open valve 103 and close valve 104.

Flush Cycle and Shut-Down

50. Re-activate the STR furnace temperature controller to 275°C (527°F).

51. Turn on the high pressure separator heaters to settings of 220 and 110 for separators 1 and 2 respectively.

52. Increase voltage to heating tapes on slurry transfer lines back to normal settings, as in actual run.

53. Close valve 102, add flush oil to V-1, open valve 102 and turn on the circulating pump.

54. Purge the high pressure metering pump with oil until the liquid is gas free. Open the plant valve and activate the high pressure metering pump.

55. Turn off the condenser cooling water.

56. When 1.5 kg of oil has been pumped into V-3, open valves 109, 110, 13 and 9. Activate the ALD system allowing it to carry through 5 cycles, then close valve 9 and allow one more cycle. De-activate the ALD system and close valves 109, 110 and 13.

57. Open valve 8 and close valve 7 to transfer oil to V-4. Close valve 20.

58. Withdraw oil from V-3. Slowly equalize pressure through valve 20.

59. After 1.0 kg of oil has been pumped into V-4, open valve 7 and close valve 8 to transfer oil to V-3. Close valve 21.

60. Withdraw oil from V-4. Slowly equalize pressure through valve 21.

61. After 1.0kg of oil has been pumped into V-3, de-activate metering pump. Close valves 7 and 20 and plant valve.

62. Shut off all heating tapes, heaters and the STR furnace.

63. Transfer contents of V-2 into an evacuated high pressure vessel through valve 6. After 15 minutes close valve 6.

64. Withdraw oil from V-3.

65. Turn off the magnedrive stirrers.

66. Drain flush oil from V-1.

67. Open valves 20 and 21 to equalize the pressure in V-3, V-4 and V-13.

POST-RUN OPERATIONS

1. Open valve 2 and pressurize the STR to 10.34 MPa (1500 psig) and then close valve 2.

2. Transfer contents of V-2 into an evacuated high pressure vessel through valve 6. After 15 minutes close valve 6.

3. Open valve 22 and pressurize V-3, V-4 and V-13 to 10.34 MPa (1500 psig) and then close valve 22.

4. Close valves 20 and 21 and withdraw residual flush oil from V-3 and V-4.

5. Evacuate the system by following Steps 1 to 6 on page G-2.

FLOWSHEET FOR CONTINUOUS COAL LIQUEFACTION UNIT